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BETA-LAYERING OF SOLID DEUTERIUM-TRITIUM IN A SPHERICAL POLYCARBONATE SHELL

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ABSTRACT

We have examined two of the variables that affect the beta-layering process in which nonuniform layers of solid deuterium-tritium (DT) are driven toward uniformity by beta-decay induced sublimation. For these experiments, a 9 mm diameter polycarbonate sphere was partially filled with a 50-50 mix of DT liquid, frozen, and then held at 17 K. We measured the equilibration time constant τ as functions of solid layer thickness, ^{4}He exchange gas pressure, and age. Solid layer thicknesses ranged from 200 μm to 650 μm, exchange gas pressures from 0 to 600 torr, and age from 0 to 104 days. Results show a significant final solid layer anisotropy with exchange gas pressures above 5 torr, and τ values that increased with age by 0.01 min/day for 200 μm-thick layers, and by 0.5 min/day for 650 μm-thick layers. The time constant is shown to be a weak function of exchange gas pressure.

INTRODUCTION

The development of high-gain targets for inertial confinement fusion (ICF) requires that the DT fuel layers exhibit a highly uniform profile. It has been demonstrated elsewhere[1,2,3] that a nonuniform layer of solid DT can be driven toward uniformity by the process of radioactively-induced sublimation (beta-layering). The tritium beta-decay produces temperature gradients[1] across the solid fuel layers which drive the sublimation of solid DT from thicker to thinner layers, forcing the layers toward uniformity. In previous work, the beta-layering process was studied in cylindrical geometry for both solid tritium[1] and solid DT layers[3,4], inside a high thermal conductivity copper cell. The cell wall interior boundary was therefore nearly perfectly isothermal, resulting in very low values of the final solid layer anisotropy δ_∞. In contrast, the present experiments utilize a low thermal conductivity spherical cell, to simulate the low-z plastic material and the actual geometry of prototype ICF reactor targets. If the thermal gradients in the ^{4}He exchange gas are not spherically symmetric, then observable values of δ_∞ could arise, with detrimental consequences to the ultimate target yield.

Aging implies simply the buildup of stagnant ^{3}He gas in the central vapor space, creating increasing resistance to DT vapor transport and thus causing the equilibration time constant τ to increase with time[4]. Using theoretical modelling, Giedt[5] has shown that when ^{3}He is present, then τ is also a function of both the cell wall thermal conductivity k_w and the heat transfer coefficient of the ^{4}He exchange gas h, which is pressure dependant.

EXPERIMENTAL SET-UP AND PROCEDURES

A detailed description of the experimental apparatus is given elsewhere[1,3], and will not be repeated here. The major components are as follows: the DT gas handling system, including a palladium bed for storage and cleaning of the DT between experiments; a closed-cycle helium refrigerator-cooled cryostat; a multilayer containment system, providing tritium containment and a cryogenic environment for the DT target cell; a polycarbonate (PC) target cell with a stainless steel fill tube; and an imaging system including an incandescent backlighter, relaying optics, a CCD camera and a video frame grabber and image processing software. Data reduction and analysis were done using a 386 computer and several high-level software packages.

The PC cell is enclosed by a secondary containment cylinder, which is filled with ^{4}He exchange gas at the desired pressure. Experiments were begun by filling the cell to the required level with DT liquid at 20 K, then quickly lowering the cell temperature to 17 K, about 2.8 K below the triple point. Figure 1. shows the sphere just after the initial freeze. Part of the DT liquid remains in the stainless steel fill tube and also freezes, providing a solid plug which prevents any further transport of DT into or out of the cell. We developed a system to correlate initial liquid level to final uniform solid layer thickness d, based on the volume of the liquid relative to the volume of the cell. Between most experimental runs, the solid

Fig. 1. A photograph of the clear spherical target cell filled ~20 % full with solid DT. This photo shows the cell just after the initial freeze, when the solid layer anisotropy is at its maximum. The solid is seen as an irregular clear (white) layer distributed along the bottom half of the interior of the cell. The stainless steel filling tube is near the bottom of the cell. A similar tube on the other side serves only to help in mounting the cell.

Fig. 2. This photo shows the nearly perfectly uniform solid DT layer that results from beta-layering. The solid shows itself here as the inner clear (white) band. The central clear circular area is an out-of-focus image of the opal diffuser mounted behind the target in the secondary containment cylinder. The same diffuser shows up as the outermost clear band, now unaffected by the spherical optics of the target shell, but still out of focus.

DT was liquified by shining an incandescent bulb into an orthogonal viewing port. This simple method maintained a constant fuel level and hence constant d from run to run because the solid plug in the DT fill tube remained frozen. Equilibration time constant, τ, and anisotropy, $\delta(t)$, were obtained by the following techniques. An image of a solid layer section was captured after focusing the optics at the vertical plane passing through the center of the PC shell (see fig. 1). With the aid of an edge-tracing software routine, ~600 points were determined around the circumference at the inner wall of the PC shell (the shell wall - DT solid interface). The precise center of the PC shell was determined using a circle-fitting routine. The average value of the internal radius of the shell, R_i, then could be determined. Another series of points were defined along the inside of the solid layer (the DT solid - vapor interface). Thus the radial distance between the center of the shell and the inner edge of the DT solid, as well as the thickness $d(\varphi)$ of the solid layer itself (by subtracting these values from R_i), was defined at 600 points along the 2π section. For a uniform solid layer, $\delta = 0$, which means that $d(\varphi)$, as measured from the center of the sphere, is constant for $0 \leq \varphi \leq 2\pi$. For a non-uniform layer, $\delta(t)$ can be determined by taking the average value of $d(\varphi)$, then determining its standard deviation, or by doing a Fourier transform of $d(\varphi)$ as a function of φ. Images of the layering process were captured at regular time intervals during the equilibration runs. Figure 2 shows the PC sphere with the DT solid layer following equilibration. The values of τ and δ_∞ for each equilibration run were determined by taking the measured $\delta(t)$ value for each time t and fitting the data to the general equation:

$$\delta(t) - \delta_\infty = \delta_0 \, e^{-t/\tau} \qquad (1)$$

where: $\delta(t)$ = instantaneous solid layer anisotropy
δ_0 = initial anisotropy
t = time from initial freeze

EXPERIMENTS AND RESULTS

1. Anisotropy v.s. exchange gas pressure

In our first set of experiments, we measured δ_∞ as the exchange gas pressure was varied from 0.2 torr to 164 torr. The cell was filled with enough DT to yield a solid layer thickness of about 500 μm, and the cell temperature was maintained at 17.6 k. The data for these experiments is shown in fig. 3. This graph shows large asymmetries at the higher exchange gas pressures, assumed to be the result of thermal convection in the ^{4}He exchange gas. As the pressure was reduced to 5 torr, the final asymmetry diminished to about 6% where it leveled off. This residual asymmetry is probably due to the effects of the metal fill tube and support tube attached to the poles of the PC sphere. It could also be due to aberrations in the optical system or the limiting resolution of our CCD imaging system (640 x 480 pixel array).

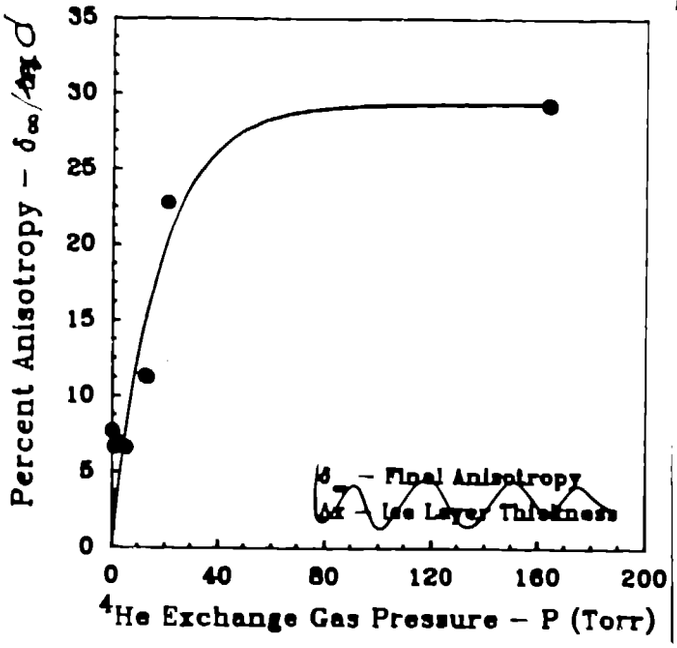

Fig. 3. Final solid layer anisotropy, δ_∞, expressed as a percentage of the total average thickness d, as a function of the ^{4}He exchange gas pressure. For these experiments, d was about 650 μm. The residual anisotropy at low pressure could be the result of heat-sinking effects of the cell fill and support tubes, aberrations in the optical system, or the limiting resolution of the imaging system.

2. Aging effect on the equilibration time constant

In the next series of experiments, shown in fig. 4, we measured the effect of DT age on τ for two separate solid layer thicknesses. The curve for the 200 μm thick layer appeared relatively flat for the first 30 days, so we did not extend these experiments. The curve for the 650 μm layer began to show an observable rate of increase after 30 days so we elected to continue this experiment to 104 days in order to quantify the nature and size of the variation. The results of the latter experiment show a nearly linear increase in τ of about 0.5-0.6 min per day. This is much lower than the 12 min/day measured previously[3] for the high conductivity cylindrical copper cell. This is due to the differences in thermal shunting between high and low conductivity cell walls. When ^{3}He is present in the vapor space, DT mass transport occurs by diffusion[6] and a temperature gradient proportional to the concentration of ^{3}He is established. This is accompanied by a DT partial pressure gradient across the vapor space which is larger than would exist if no ^{3}He were present. In effect, the system responds to the impedance of ^{3}He by building up larger temperature and pressure gradients, and the overall rate is only slightly affected. However, in high conductivity cells like the copper cylinder, the temperature gradient in the vapor space is partially shunted at the wall, reducing the equilibration rate drastically[5]. Additionally, the ^{3}He buildup rate in the vapor space was somewhat larger for the copper cylinder than for the spherical target because of the larger fraction of DT solid to cell volume in the cylinder experiments[1,3,4]. This effect is also responsible for the difference between the two sets of data in fig. 4.

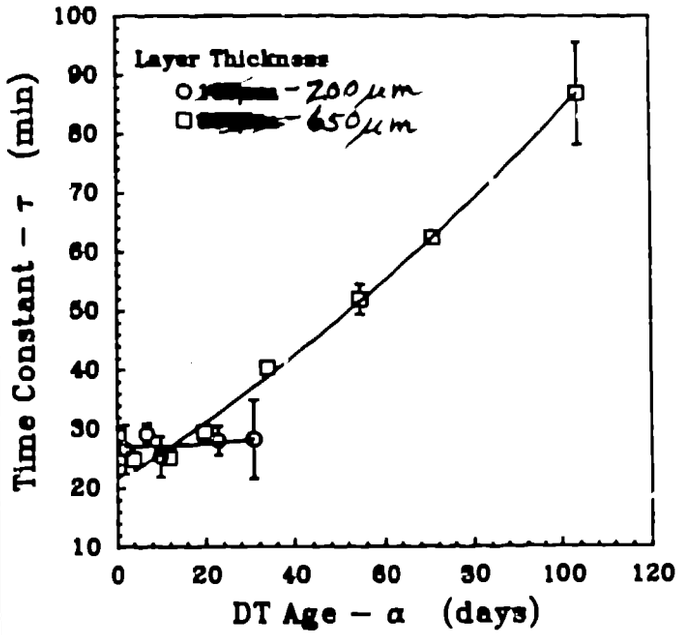

Fig. 4. The equilibration time constant τ vs. the age α of the DT fuel in the target. Aging causes a buildup of stagnant ^{3}He gas in the central vapor space, which tends to impede the mass transfer of DT. The effect is more noticeable for the 650 μm thick layer, where the overall rate of ^{3}He production is considerably faster. Each data set was fit to a second order polynomial with the results: $\tau = 27 - .0092\ \alpha + .0016\ \alpha^2$ (200 μm thickness), and $\tau = 21 + .477\ \alpha + .0015\ \alpha^2$ (650 μm thickness).

3. Time constant vs. exchange gas pressure

A one-dimensional analysis of the beta-layering process in spherical geometries was performed by Giedt[5]. He examined the effects of varying the DT cell wall thermal conductivity and the ^{4}He exchange gas pressure on the equilibration time constant. We performed a set of experiments to measure the latter effect. These experiments were done by removing the ^{3}He from the DT between each run and refilling the Lexan sphere, producing "fresh" samples for each run. The exchange gas pressure was set to the desired value before each fill

and equilibration run. The ^{4}He pressures available to us limited the maximum h value to about 4.5. The results are shown in fig. 5., along with the theoretical prediction made by Giedt. This graph is a rearrangement of fig. 8. of Ref. 5 with ^{4}He exchange gas pressure converted to h. It shows the effects of h on τ for several values of wall thermal conductivity. Our data are for a PC sphere with a 650 μm solid layer, and were taken at zero sample age. The Giedt curves are for slightly thinner solid layers (500 μm) and were generated for a sample age of ten days. Our data do fall within the range expected for the wall conductivity and sample age used and track the slope of the Giedt's predictions nicely. However, Giedt did not consider the case of zero age, where the curves in Fig. 5, might all collapse to the ideal minimum rate constant of 26.6 minutes[1]. Thus our data show only that τ is a relatively weak function of the ^{4}He exchange gas pressure when no ^{3}He is present.

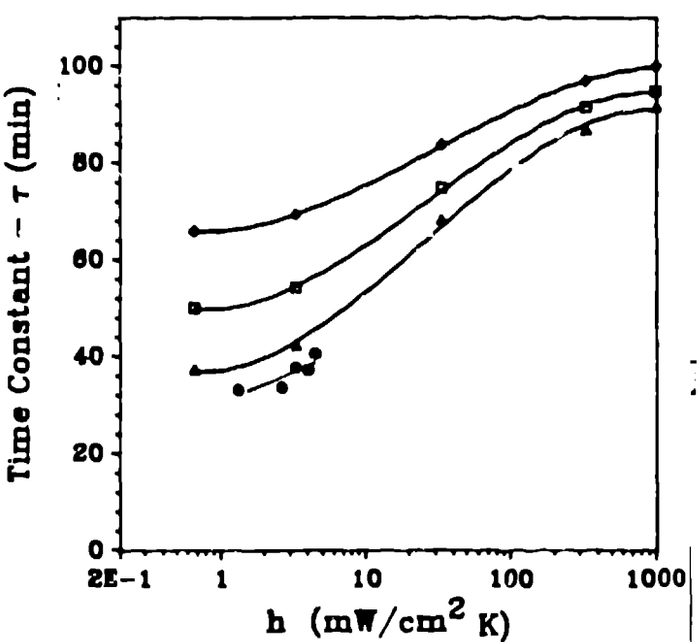

Fig. 5. The equilibration time constant τ as a function of the ^{4}He exchange gas pressure (converted to the equivalent heat transfer coefficient h to compare with the predictions of Giedt[5]). Giedt's calculations (open symbols) are for a 10 day old sample and a 500 μm thick solid DT layer, while our data (closed circles) are for nearly fresh DT (zero age) and a 650 μm layer thickness. For the three predicted curves, target wall thermal conductivity increases vertically: Δ - 0.001 W/cm K, $\square$ - 0.01 W/cm K, and $\diamond$ - 0.03 W/cm K.

CONCLUSIONS

There are striking differences in the details of beta-layering occurring within low-conductivity polycarbonate walls, as opposed to high-conductivity copper walls as observed previously[1,3,4]. The most noticeable effect is the large amount of final anisotropy. This effect decreases as the pressure in the ^{4}He exchange gas is lowered, reaching a minimum below ~5 torr, suggesting that it is due to thermal convection. Maintaining adequate cooling with purely conductive heat transport (which occurs at pressures below ~2 torr) should not present a problem for the target sizes of interest to ICF applications. However, whether the final anisotropy can be reduced to meet the stringent conditions required for laser-driven implosions is still an open question.

The effect of DT age (^{3}He content) on the equilibration rate in a polycarbonate shell is almost non-existent, in contrast to the strong effect observed in copper. For practical ICF targets, the effect will not be significant unless the DT is very old.

Finally, the fact that these observations are in excellent agreement with computer modelling predictions by Giedt[5] and others[2,6] shows that our theoretical understanding of the beta-layering phenomenon is adequate.

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