

Role of Hydrogen Fractionation in ICF Ignition Target Designs

Introduction

The need of using cryogenic hydrogenic fuels in inertial confinement fusion (ICF) ignition targets has been well established. Efficient implosion of such targets has mandated keeping the adiabat of the main fuel layer at low levels to ensure drive energies are kept at a reasonable minimum. In fact, it has been shown by many authors that the minimum drive energy of an ICF implosion scales roughly as the square of the fuel adiabat.¹⁻³ The use of cryogenic fuels helps meet this requirement and has therefore become the standard in most ICF ignition designs.

To date, most theoretical ICF ignition target designs have assumed a homogenous layer of deuterium-tritium (DT) fuel kept roughly at or just below the triple point. Such assumptions have led to several promising ICF target designs⁴⁻⁷ that have numerically demonstrated ignition and burn under a variety of illumination schemes. Recent work done at the Laboratory for Laser Energetics (LLE), however, has indicated the possibility that, as cryogenic fuel layers are formed inside an ICF capsule, isotopic dissociation of the tritium (T), deuterium (D), and DT can take place, leading to a “fractionation” of the final ice layer.

Fractionation, as illustrated in Fig. 105.35, can lead to isolated areas of the ice layer that are either T rich or D rich. Under such circumstances, the performance and overall viability of previous ignition designs need to be examined.

Motivation

The possibility of isotopic dissociation leading to fractionation within cryogenic hydrogenic fusion fuels was first proposed by Prigogine⁸ with initial experimental investigations carried out by Bienfait.^{9,10} It is surmised that, because of the different triple point temperatures of the various constituent molecules (T₂, D₂, and DT), as a cryogenic layer is formed, the individual constituents freeze-out separately from the whole. As shown in Fig. 105.35, such a situation could lead to large volumes of the cryogenic layer being totally devoid of DT molecules. The significance of this can be understood when examining the thermonuclear reaction rates for these three molecules as a function of temperature, as shown in Fig. 105.36. The expected operating temperature for inertial confinement ignition target designs, as they approach the ignition threshold, is typically held to be in the 8- to 12-keV range.¹¹ As can be seen in Fig. 105.36, the reaction rates for both the T + T and

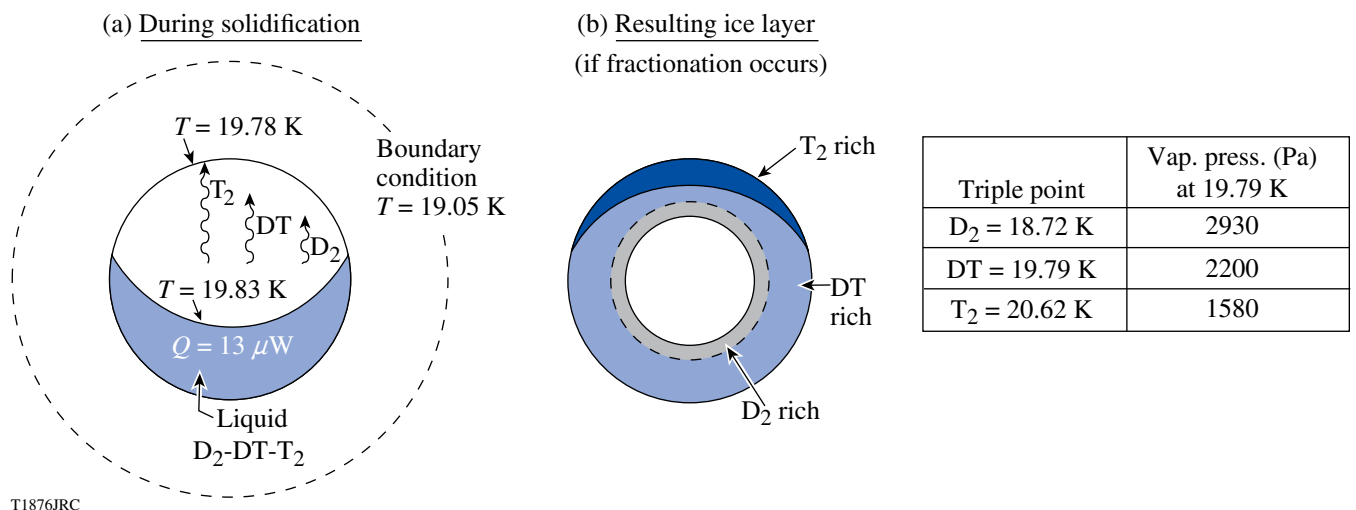


Figure 105.35

(a) Fractionation of isotopic hydrogen mixtures can occur because of the variance in the triple point. (b) Such fractionation could lead to isolated fuel layers.

D + D reactions are several orders of magnitude lower than that for the D + T reaction.¹²

Several initial scenarios of fractionation, as illustrated in Fig. 105.35(b), considered complete separation of the constituents into unique and isolated regions of the cryogenic layer. The overall effect of the fractionation on target performance is then dependent on the preponderance and spatial deposition of the separated fusion-fuel molecules. Of particular note is the scenario in which the D₂ molecules are the very last species to solidify. Such a scenario is illustrated in Fig. 105.37(a), where a solid D₂ crust has formed on the inner surface of the ice layer. This is particularly damaging in that when a normal ignition experiment is assembling in preparation for ignition, a central hot spot is required to form at the very center of the target. The majority of the material in this hot spot is made up from the mass from the inner few microns of the ice layer, which, because of the large temperature present at the center due to viscous work, has ablated into the region. As the ablated material is heated, the need for large concentrations of fusing DT molecules and their fusion by-product alpha particles for the initiation of the ignition process is obvious.

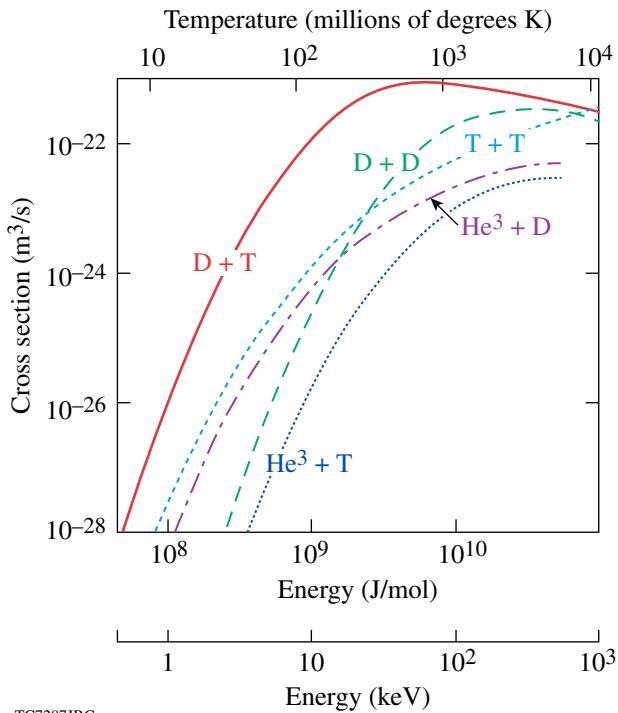


Figure 105.36
Thermonuclear cross sections for the five most easily activated fusion reactions indicate that DT fusion is the most easily obtained for ICF implosions. (Figure courtesy of the University of California/Lawrence Livermore National Laboratory.¹²)

The effect of such a scenario on target performance can be quickly examined using the 1-D radiation-hydrodynamic computer code *LILAC*.¹³ Several ignition implosion simulations were considered in which the inner surface of cryogenic DT fuel was replaced by solid D₂, as illustrated in Fig. 105.37(a). As shown in Fig. 105.37(b), as this inner layer increases in depth, target performance begins to degrade until eventually the target fails to achieve ignition. This behavior is due to the increasingly DT-depleted fuel that is being ablated into the hot-spot region, failing to produce the necessary alpha particle deposition in the cold, dense fuel surrounding the hot spot.

Another aspect of complete fractionation is the possible formation of a polar cap of pure T₂. Simulations of such scenarios require the implementation of a 2-D radiation-hydrodynamics computer code such as *DRACO*.^{14,15} An extreme example of the effect of complete polar separation is given in Fig. 105.38(a).

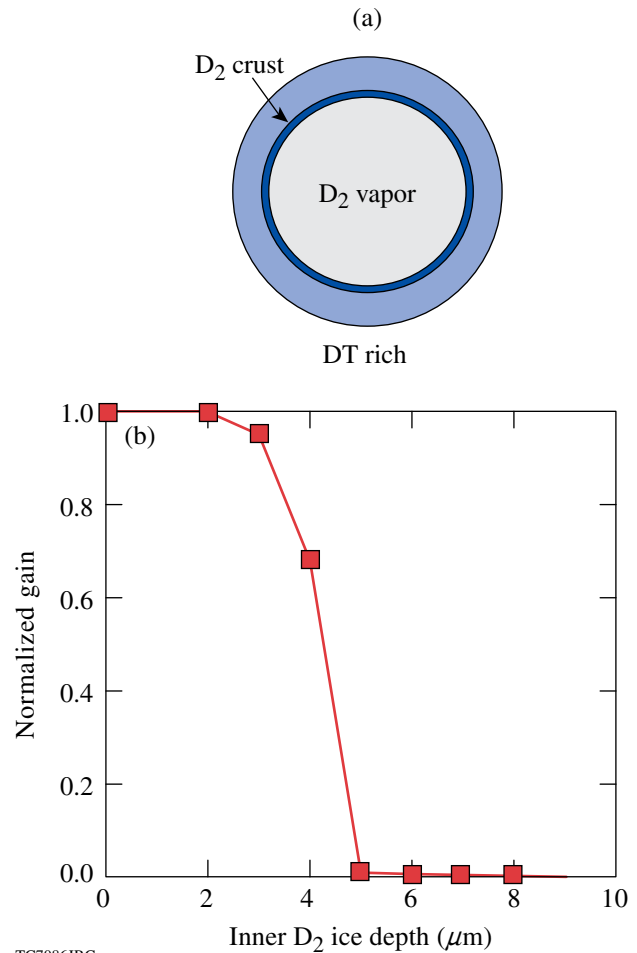


Figure 105.37
(a) Complete inner fractionation denies the ignition hot spot of the necessary tritium, which, (b) in abstentia, can preclude ignition.

Here we consider not only the formation of a northern pole cap of T_2 but, in addition, assume that a southern pole of pure D_2 has also formed. An isodensity plot of this target implosion taken at a time in the experiment after the thermonuclear blast wave has moved through the DT-rich part of the target in the equatorial regions is shown in Fig. 105.38(b). What is apparent in the density plot is that the polar regions of the target have not been able to participate in the burn because of the lack of DT. As such, while the burn wave has severely decompressed the equatorial regions, the polar caps have remained at very high densities because of the compressional nature of the ICF implosion. The overall performance of this design has suffered dramatically with the gain dropping from 45 to about 15.

Results such as these clearly indicate the need to experimentally determine the possibility and overall extent of hydrogenic fractionation in ICF ignition capsules.

Experimental Setup

An experimental test bed to investigate the isotopic fractionation of fusion fuels was designed, built, and implemented at LLE, as illustrated in Fig. 105.39. The system was designed for use with nonradioactive isotopes and, as such, only examines fractionation scenarios involving $H_2:HD:D_2$ mixtures. We have investigated the possibility of fractionation using a deuterium–hydrogen ($H-D$) mixture with a similar isotopic ratio to the $D-T$ mixture to be used in future cryogenic experiments

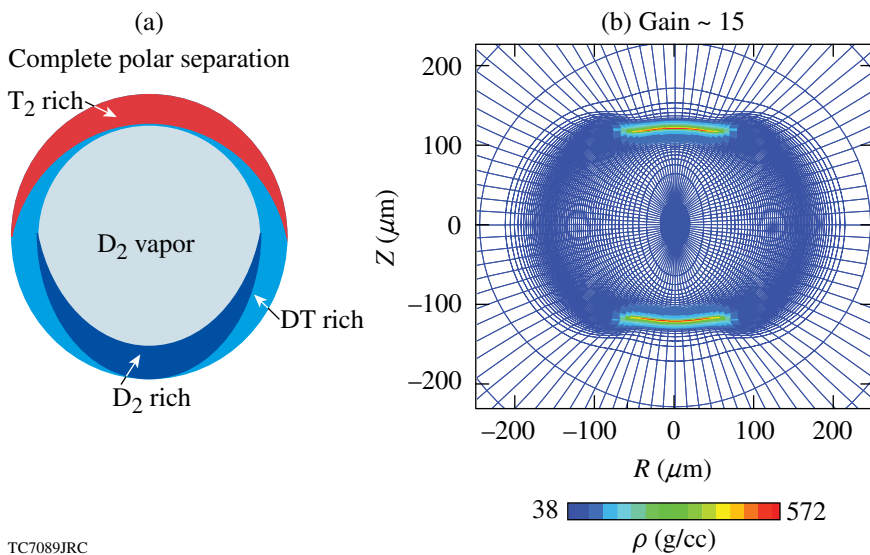


Figure 105.38
 (a) A polar cap fractionation scenario exempts the DT-poor poles from the ignited burn, reducing the performance of a high-gain implosion.
 (b) High-density polar regions are evidence of nonburning fuel.

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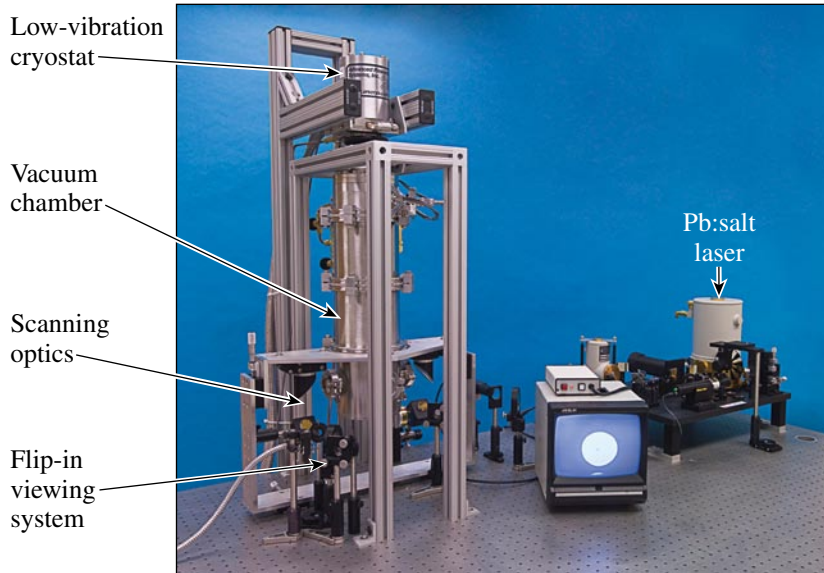


Figure 105.39
 A $H_2/HD/D_2$ fractionation test bed is used to measure the IR absorption coefficient in a cryogenically solidified mixture.

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(25% H₂, 50% HD, and 25% D₂) along with a slow-freezing protocol (–10 mK/h over a ~1-K range) that has been demonstrated to produce good-quality targets using the OMEGA Cryogenic Target Handling System. A focused beam from a Pb:salt laser tuned to the 3162 cm⁻¹ absorption band of D₂ is raster scanned across a 6-mm-diam, 3-mm-thick sample of the mixture to determine the D₂ concentration as a function of position. This process was performed on samples that were solidified over several hours to several days to look for differences in fractionation due to diffusion of the different molecules to the liquid/solid interface as the sample was cooled.

Several mixtures of various H₂:D₂ ratios were examined in addition to the 25:50:25% H₂:HD:D₂ mixture. By plotting the absorption coefficient of the D₂ in the H–D as a function of the D₂ fraction in the mixture, the slope can be used to quantify a transmission gradient as a concentration gradient. The absorption coefficient for D₂ in a H–D mixture was found to be exponentially dependent on the D₂ molecular fraction in the mixture, as illustrated in Fig. 105.40. From this, a maximum D₂ concentration gradient of 0.02 to 0.05 molecular fraction per millimeter was observed for the samples, as shown in Fig. 105.41. The average D₂ concentration gradient (percentage) is greatest for the 25% molecule fraction of D₂, which is representative of the 25:50:25% H₂:HD:D₂ mixture. The large error bars shown for the lowest concentrations indicate the signal-to-noise ratio in the measurement is smallest when the sample is the least absorptive.

Experimental Results

A complete and thorough discussion of the experimental results are presented earlier in this issue in **Isotopic Fractionation During Solidification of H₂–HD–D₂ Mixtures**

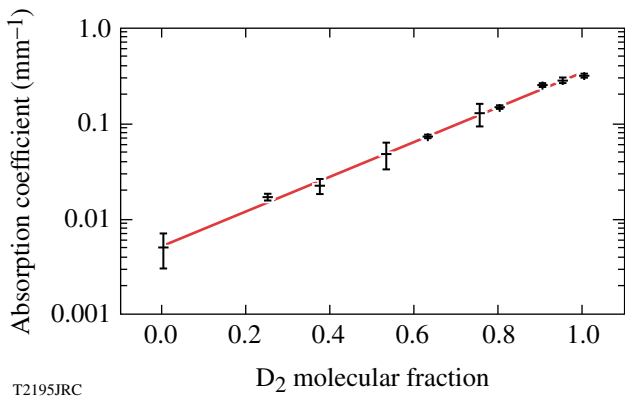


Figure 105.40
The average absorption coefficient of the D₂ in a H₂–D₂ mixture was measured as a function of the D₂ molecular fraction.

on pp. 29–32. We have included a subset of this discussion to establish the relevant experimental initial conditions needed in the numerical modeling of fractionation.

Initial experiments focused on answering the question of whether or not complete fractionation, the situation where regions of pure, single isotopes freeze-out separately, occurs for these types of isotopes. Of particular interest was the experimentally observed, “first-freezing” (FF) temperature of a given cryogenic sample. The FF temperature is the temperature of the cryostat at which definitive crystal growth is observed. For pure samples or cases of complete fractionation, this would, of course, be the normal triple point of the material as given in Table 105.I on p. 32. Also given in the table are approximations for the FF temperature based on various models for the mass fraction of a given mixture. The FF temperature is calculated from a weighted sum of the product of the assumed isotopic mass fraction f_i and its appropriate triple point temperature $T_{tp,i}$ as given by

$$\sum_i f_i \times T_{tp,i}$$

From this approximation one can see that the FF temperature for this mixture should lie in the range from 15.72 to 16.49 K. Additionally, the entire mixture should freeze-out into a layer when the FF temperature is reached and held constant.

The experiment was conducted and attention was focused on the mixture as the cryostat temperature approached the first triple point of the mixture constituents (deuterium) at 18.73 K. No appreciable crystal growth was observed. The same was true when the cryostat temperature reached and fell below the triple point of the HD molecule at 16.60 K. This experiment

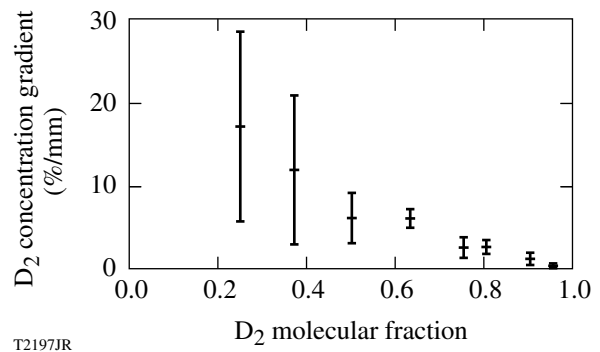


Figure 105.41
The average D₂ concentration gradient (percentage) is greatest for the lowest concentrations.

was repeated several times with the same results and indicates the absence of complete fractionation in cryogenic fusion fuels. As the experiment proceeded, it was only at a temperature of 16.53 K that appreciable crystal growth appeared. However, the entire sample did not freeze-out into a layer as the temperature was held constant. It was only after the cryostat temperature was further lowered to 16.10 K that total solidification of the mixture was observed. While the results from these experiments indicated that complete fractionation of cryogenic fuel layers does not take place, the differential temperatures required to freeze the entire sample did indicate the presence of low levels of fractionation within the mixture.

Additional experiments that examined the spatial dependence of the IR transmission coefficient of a cryogenic sample layered within the low-vibration cryostat were then carried out. As illustrated in Fig. 105.42, the experimental system recorded a spatial variation in the transmission coefficient of the cryogenic sample. Figure 105.42 also shows the singular transmission levels for pure H_2 and D_2 samples. From this image it is clear that fractionation has occurred within the cryogenic fusion-fuel layer. Interpretation of the variance in transmission indicates that fractionation levels in the sample are in excess of 5% from one side of the cell to the other and could in fact be as high as 10% overall.

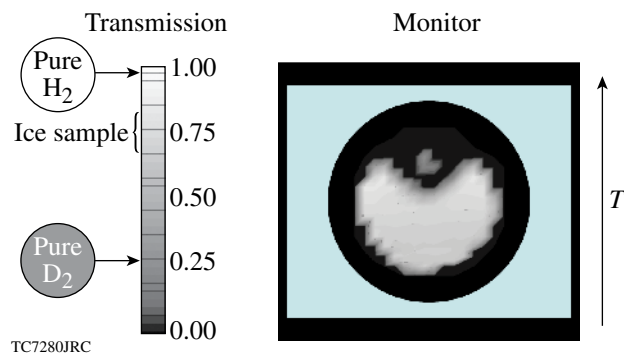


Figure 105.42
The absorption coefficient of the D_2 in the H/D mixture is less than 1/20th of that for pure D_2 .

Numerical Results

A numerical fractionation scenario based on the above experimental results was constructed and studied. The particular metric investigated was the effect of the fractionation on target gain. Using the template illustrated in Fig. 105.43, the north polar fractionation within the target was numerically varied up to levels of 100%. As the level of fractionation was increased past ~30%, the increasing lack of DT at the poles

of the target began disrupting the target performance. As can be seen in Fig. 105.44, this process can degrade target performance from a gain of 45 for perfect ice down to less than 10 for the cases with high levels of fractionation. For small levels of fractionation, the ignition and burn phases of the implosion proceed almost completely unaffected by the redistribution of the fusion fuel within the target. It is helpful to remember that in most ICF ignition designs the burnup fraction of fusion fuel is typically only in the 10%–15% range by molecule. As such, the target performance is relatively unaffected by small changes in the distribution of the fusion-fuel molecules. As was discussed earlier, however, as the amount of DT within a specific volume of the target is reduced, the likelihood of that region participating in the ignition burn wave is greatly diminished.

Another indication of how increasing fractionation perturbs target performance can be seen from a comparison of the

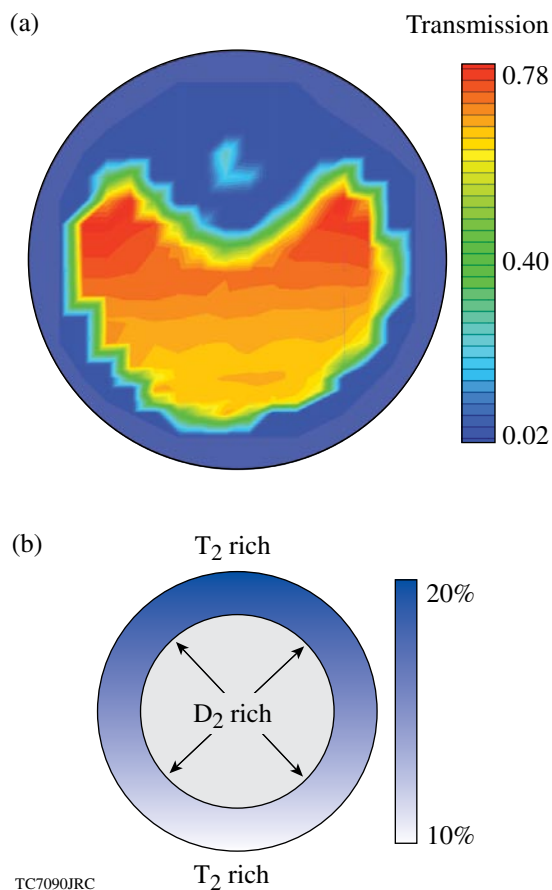
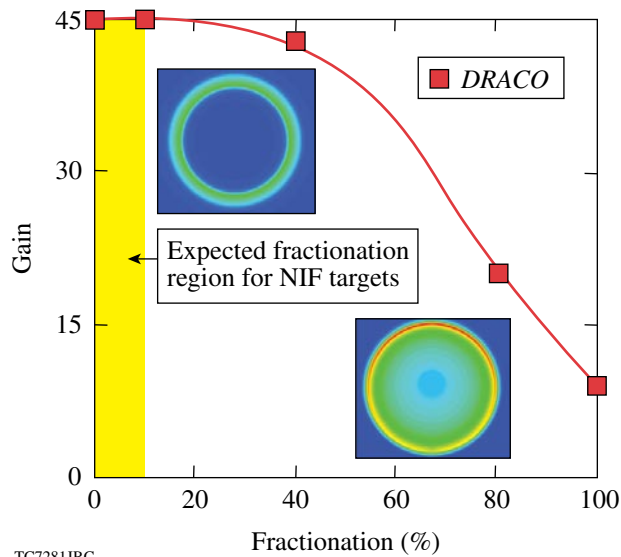


Figure 105.43
(a) Isotopic hydrogen ($H_2/HD/D_2$) fractionation in a solution has been observed in the laboratory at levels approaching 10%. (b) This information was used to construct a set of initial conditions for numerical studies of fractionation in ICF target designs.



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Figure 105.44

Fractionation levels in excess of ~30% are required before ignition target performance is affected. Current estimates place a 10% upper limit on the fractionation—too low to degrade target performance.

isodensity contour plots given as insets in Fig. 105.44. These contours, drawn on the same legend, demonstrate the symmetric decompression of the ice layer in the low fractionation cases. While similar decompression is apparent in the lower hemisphere of the high-fractionation case, the north polar region has remained at high compressed densities because of the region's inability to participate in the DT thermonuclear burn.

While the effects of high levels of fractionation on target performance are disturbing, the good news is that, given the experimental evidence to date, current estimates of the levels of fractionation in NIF ignition target designs are not expected to exceed 10%. As such, fractionation is not viewed as a major threat to the overall performance of ICF ignition design planned for experiments on the NIF, as illustrated as the shaded region of Fig. 105.44.

Conclusion

The need of using cryogenic hydrogenic fuels in ICF ignition target designs has been well established. Efficient implosion of such targets has mandated keeping the adiabat of the main fuel layer at low levels to ensure drive energies are kept at a reasonable minimum. To date, most theoretical ICF ignition target designs have assumed a homogenous layer of DT fuel kept roughly at or just below the triple point. However, recent work done at LLE has indicated the possibility that, as cryogenic fuel layers are formed inside an ICF capsule, isotopic dissociation of the T, D, and DT can take place, leading to a “fractionation”

of the final ice layer. Fractionation studies of fusion-like fuels (H/HD/D) have demonstrated the existence of fractionation in ICF cryogenic fuel layers. However, numerical simulations of ignition target designs, using experimental fractionation scenarios, indicate that small levels of fractionation (~10%) are acceptable for ignition performance on the NIF.

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