

Current Status and Issues in Tritium Technology to Produce Laser Fusion Target

T. Norimatsu, M. Takagi and K. Mima
Institute of Laser Engineering, Osaka University
2-6, Yamada-oka, Suita, Osaka 565, Japan

Current status of tritium technology to fabricate laser fusion targets is described. Tritium facilities for laser irradiation, deuterium-tritium (DT) fuel loading system, fabrication of deuterated-tritiated polystyrene shells, measurement of partial pressure of tritium and cryogenic technologies to fabricate a thick solid DT layer are briefly mentioned. Issues to fabricate cryogenic DT target for the coming upgraded laser system is also discussed.

1. Introduction

After achieving high temperature[1] and high density compression[2], laser fusion research proceeded into a new phase to demonstrate ignition and energy gain.

At Institute of Laser Engineering, Osaka University, two approaches are examined to show the ignition using the new GEKKO XII laser system. One is a conventional implosion with a central ignitor in which the main fuel is burnt by alpha-particles from the central ignitor. In this scheme, spherical symmetry of the implosion process is the key issue for success. The other is the fast ignition in which compressed fuel is fired directly by an ultra-short, intense laser. In this scheme, the energy deposition into the compressed core is our interest.

A highly spherical, cryogenic target with a uniform solid deuterium and tritium (DT) layer is inevitable for both implosion schemes. From a viewpoint of tritium technology, generation of 60 to 100 MPa DT gas to make the thick solid DT layer, development of a fill and transport system for the DT-filled cryogenic target, analysis of the solid DT layer inside the cryogenic target and behavior of tritium in the target chamber must be examined.

2. Status

2.1 Tritium facilities

Tritium facilities for the GEKKO XII laser system consist of a pellet factory to fabricate targets filled with a deuterium-tritium mixture and two target chambers; one is designed to use in direct-irradiation implosion with uniform beams and the other is for indirect-irradiation






implosion of cannon ball targets with two-bundled beams. The boundary of radioactive hot zone is settled on the surface of the target chamber. Tritium of 3.7×10^9 Bq can be used in a day at the target chamber.

The vacuum system of the target chamber consists of two cryogenic pumps, a turbo molecular pump and two rotary pumps. The vacuum exhaust gas from the target chambers is sent to a tritium recovery system in the pellet factory. The gas is stored in a buffer tank and circulated to remove tritium contamination using a conventional oxidizer and molecular sieves. Copper oxide was used to oxidize hydrocarbon with tritium.

Varieties of targets such as glass microballoons filled with DT gas, plastic shells made with deuterated, and tritiated polystyrene and plastic shells filled with DT gas are used in implosion experiments with the GEKKO XII laser to study volume ignition, high density compression and the central ignition, respectively. Typical specifications for these targets are summarized with future plans in Table 1. These targets are fabricated in a pellet factory capable of handling 30 TBq tritium in a day and transferred to target chambers for implosion experiments.

In the pellet factory, there are two glove boxes in which a fuel-fill station for routine targets and that for the next step are housed. The glove boxes are equipped with a tritium recovery system capable of recovering 3.7 TBq tritium in 3.5 hr with a decontamination factor of 10^5 . This tritium recovery system is also used for clean up of tritium accidentally released in the room.

Table 1 Specifications for laser fusion targets

Status/ Purpose	Specifications	Tritium in a shell	Tritium in a factory
Developed/ High temperature implosion	 Glass shell filled with gaseous DT <math>< 1 \text{ MPa}</math> Diameter, 1.2 mm Wall thickness, 1.2 μm	$3.7 \times 10^8 \text{ Bq}$	$1.5 \times 10^{13} \text{ Bq}$
Developed/ High density Implosion	 Deuterated-tritiated plastic shell Diameter, 600 μm Wall thickness, 8 μm	$3.7 \times 10^8 \text{ Bq}$	$1.5 \times 10^{13} \text{ Bq}$
Next plan/ Ignition	 Plastic shell with Solid DT layer Diameter, 1200 μm Wall thickness, 20 μm Solid DT, 60 μm in thickness	$3.7 \times 10^{10} \text{ Bq}$	$2 \times 10^{14} \text{ Bq}$
Future plan/ Energy gain	 Plastic shell with Solid DT layer Diameter, 3 mm Wall thickness, 60 μm in thickness Solid DT, 150 μm	$1 \times 10^{12} \text{ Bq}$	$2 \times 10^{15} \text{ Bq}$
Future plan/ Reactor	 Plastic shell with Solid DT layer Diameter, 5 mm Wall thickness, 100 μm Solid DT, 250 μm in thickness	$4 \times 10^{12} \text{ Bq}$	$2 \times 10^{17} \text{ Bq}$

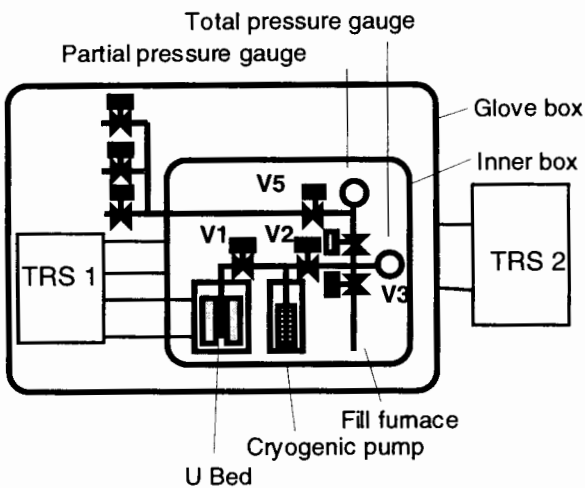


Fig. 1 Fill station for highly pressurized tritium.

The tritium-fill station for routine targets consists of a uranium bed with an equivalent DT mixture, a piezo electric transducer to measure the total pressure, a fluorescent partial pressure monitor and a fill furnace. Glass microballoons to be filled with DT fuel are housed in the furnace filled with the DT gas. The furnace is heated to 350 C for 24 hour to

allow diffusion of DT through the glass walls. To fabricate the deuterated-tritiated shell, this furnace is replaced with a quartz tube to allow irradiation of UV light.

Figure 1 shows a schematic diagram of the tritium fill station that is now being tested using deuterium. The high pressure lines of the system are built in an inner box equipped with a compact tritium recovery system (TRS-1). The inner box and the TRS-1 are, in turn, housed in a glove box equipped with TRS-2.

Tritium stored in a uranium bed is released into a cryogenic pump cooled less than 18 K. After all the tritium is condensed in the cavity, the valve 1(V1) is closed and the tritium is released into the fill furnace to produce highly pressurized DT gas by elevating the temperature of the cryostat.

The system is designed to produce high pressure using a small tritium inventory. To date, 20 MPa deuterium gas was produced using 0.027 mol deuterium (equivalent to DT gas of 30 TBq). In the near future, the fill furnace will be replaced with a portable cryostat to carry the DT-filled polystyrene shell at 18K to the target chamber.

2.2 Measurement of partial pressure of tritium

A ratio of tritium in the DT mixture is one of the important parameters in implosion experiments. We have been developing a partial pressure monitor of tritium in the fill station by using inorganic fluorescent powder[3]. A cross section of the detector is illustrated in Fig. 2. The cavity for the powder is 2.4 mm in diameter and 6 mm in length. The fluorescence is guided to a photo multiplier through a quartz rod and an optical fiber.

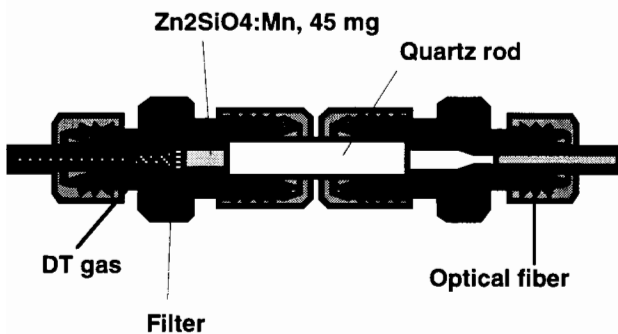


Fig. 2 Cross section of tritium partial pressure monitor.

Thirteen different fluorescent powders were examined in this experiment; hexa-ZnS:Cu(A), hexa-ZnS:Cu(B), cubic-ZnS:Cu, ZnS:Ag, ZnS:Cu:Al, ZnS:Au:Cu:Al, (ZnCd)S:Cu:Al, (ZnCd)S:Ag, Zn₂SiO₄:Mn, Y₂O₂S:Tb- Y₂SiO₄:Ce, Y₃Al₃O₁₂:Tb and Y₃Al₅O₁₂:Ce. Among these powders, the cubic-ZnS:Cu, and Zn₂SiO₄:Mn showed excellent characteristics as the detector.

When the amount of powder in the cavity exceeds 40 mg, the powder becomes optically thick and the intensity of luminance from the powder is independent of the amount of powder. The linearity of the fluorescence to the partial pressure of tritium in DT mixtures is shown in Fig. 3. Since the diameter of the powder particles are in an order of several micrometers, self-absorption of β -rays by hydrogen in the gaps between the powder particles is negligible. Although our experiments were carried out under tritium partial pressure of 1 MPa, this linearity can be extrapolated theoretically up to 10 MPa.

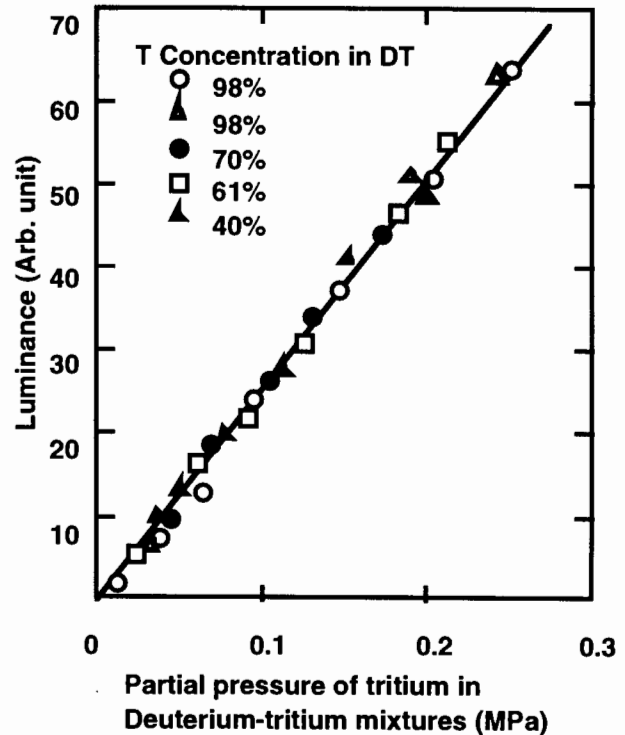


Fig. 3 Dependence of luminance on the partial pressure of tritium in DT gas measured with the Zn₂SiO₄:Mn fluorescent powder. The mixture ratios of the DT gases are shown by different symbols.

Background noise was increased and saturated within 3 repeated uses. A noise level due to the dark current of the photo multiplier was 0.06 (corresponds to 5 Pa). Saturated noise after repeated exposure was at a level of 0.3 which corresponds to the tritium partial pressure of 1 kPa. This value is negligible for use in the DT filling system.

Figure 4 shows the dependence of the signal and the background on the particle size of Zn₂SiO₄:Mn. The measurement was made with a DT gas of 52 % T at a total pressure of 0.8 MPa. The cavity in the detector was filled with the powder without any compression. The signal increased and the background decreased as the particle size became smaller. Luminance with 2.7 μ m powder was 1.24 times larger than that with 7.8 μ m powder. This ratio is very close to the ratio of the total surface areas for the 2.7 μ m and 7.8 μ m powders filled in the same volume. This good agreement is explained by the small penetration depth of β -ray into the powder particle. The ratio of the backgrounds for the 7.8 μ m powder and the 2.7 μ m powder is approximately equal to the mass ratio of the

powders filled in the cavity (35 mg for 2.7 μm powder and 80 mg for 7.8 μm powder). This result indicates that the memory effect is caused by the tritium diffused into the particles.

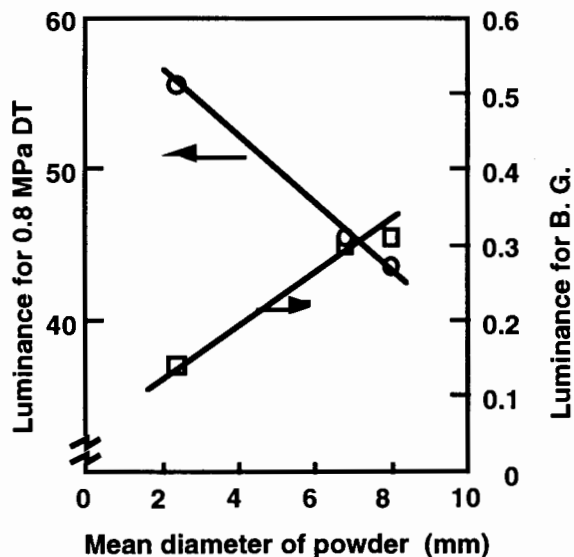


Fig. 4 Dependence of signal and noise on powder diameter. Smaller particle size is desirable for the detector.

2.3 Fabrication of deuterated-tritiated polystyrene shells

Deuterated-tritiated polystyrene shells with a silicon dopant for use in laser fusion experiments were fabricated by means of an isotope exchange method[4]. In order to increase the isotope exchange rate, deuterated polystyrene shells in a tritium atmosphere were exposed to ultraviolet (UV) radiation from a mercury lamp.

Our process starts with a synthesis of polystyrene (PS). Protium/deuterium PS [referred as HDPS, i.e., $(\text{C}_8\text{H}_4\text{D}_4)_n$] was copolymerized with equivalent normal hydrogen and deuterated PS monomers. Shells are fabricated by a density matched emulsion method described elsewhere[5]. Hydrogen atoms in the PSs are then replaced with tritium.

A cross section of the apparatus for tritium exchange is shown in Fig. 5. The apparatus consists of a quartz vessel (2 mm inner diameter, 6.3 mm outer diameter, and 150 mm length) for tritium confinement and a quartz pipe (9 mm inner diameter and 150 mm length) for water cooling. Shells were housed in a quartz capillary in a line so that they were uniformly exposed to UV light. The vessel was connected to a tritium handling system

with Swagelok joint and nylon ferrules. An equivalent DT mixture or a pure tritium gas of 0.2-2 MPa in pressure was employed to expose the DPS or HDPS shells in the quartz capillary. In order to enhance the exchange rate, a spiral low pressure mercury lamp (30 mm in loop diameter, 12 mm in tube diameter and 70 W in electric input power) was set outside the quartz tube to irradiate PS shells with UV light.

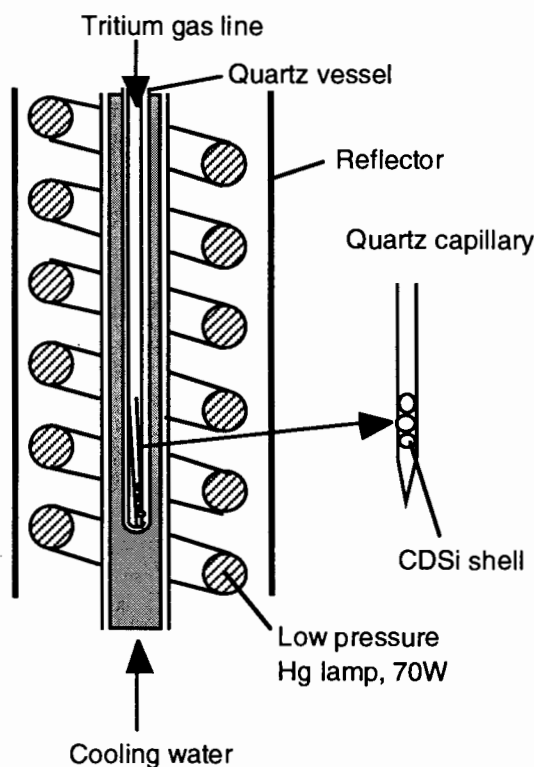


Fig. 5 Schematics of the apparatus for tritium exchange at room temperature.

An auto radiography was employed to analyze the distribution of tritium in the shell. A DPS sphere in a tritium atmosphere was irradiated with UV radiation in the same way. The DPS sphere was then molded in an epoxy resin and lapped until the cross section of the sphere appeared on the mold surface. The hemisphere in the mold was put on an auto radiographic emulsion and exposed the emulsion to β rays from tritium decay.

The experimental results are summarized in Fig. 6. The specific activity of tritium in the shell is proportional to the exposure time of UV light and no relationship with the concentration of Si dopant as a plasma diagnostic tool. The specific activity of tritium in the shells was 3.4 TBq/g for partially deuterated polystyrene and 5.6 TBq/g for fully

deuterated polystyrene, respectively. The isotope exchange rate with UV irradiation was 230 times as fast as that without the UV irradiation.

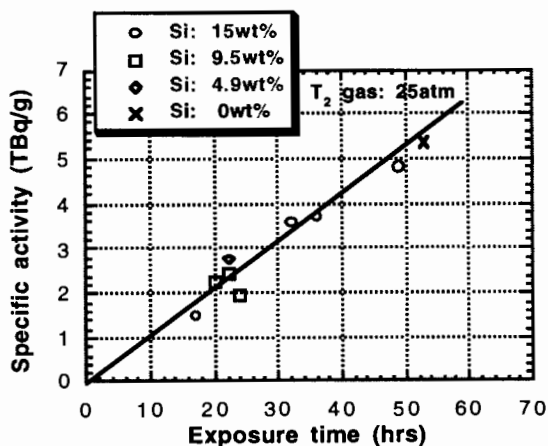


Fig. 6 Specific activity of tritium after exposure of UV light in 2.5 Mpa T_2 gas.

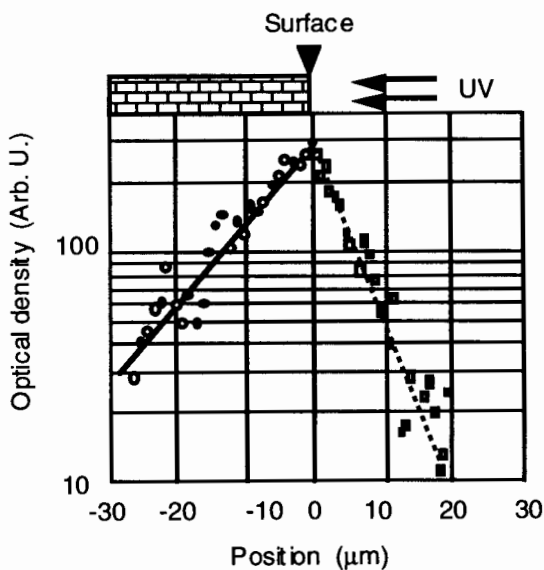


Fig. 7 Tritium distribution obtained by contact auto radiography of the cross section of shell.

The tritium in the shell was found to be strongly localized at the outer surface. Figure 7 shows distribution of tritium in the shell. The solid line for the position $< 0 \mu\text{m}$ is obtained by calculation based on reported transmittance of UV light. Distribution of tritium for position $> 0 \mu\text{m}$ is attributed to the

diffusion of tritium into the mold during polishing process.

2.4 Fabrication of polystyrene shell with long fuel retention

Recently implosion experiments have been carried out using polystyrene shells instead of glass microballoons since the glass microballoon causes preheat of fuel due to x-rays. Polystyrene shells, however, cannot keep D_2 or DT gas for its high permeability ($7.2 \times 10^{-15} \text{ mol m/m}^2 \text{ sec Pa}$) at room temperature. We have started to fabricate polystyrene shells over-coated with an Al layer to keep D_2 or DT gas. The influence of the Al coating on the preheat is negligible. This technique was originally developed by M. D. Wittman et al. at University of Rochester[6].

The experimental results are shown in Fig. 8. When the 81-nm-thick Al layers are directly coated on 700 to 800 μm -diameter polystyrene shells with electron beam vapor deposition, the apparent permeability of D_2 gas was $1.01 \times 10^{-18} \text{ mol m/m}^2 \text{ sec Pa}$ at room temperature. This permeability was too large to use the shell in an implosion experiment at room temperature.

When a 30 to 97 nm thick Al coating was applied to a polystyrene shell that has been precoated with a 0.5-1.5 μm thick parylene layer, the permeabilities of D_2 and DT gas were 1.5×10^{-20} , $1.1 \times 10^{-20} \text{ mol m/m}^2 \text{ sec Pa}$ at 23 C, respectively. The precoating of parylene N was effective to reduce the apparent permeability.

Aluminum layers were also coated by a sputtering method on the surface of shells precoated with parylene N layers. The permeability of DT gas at 23 C was $1.1 \times 10^{-20} \text{ mol m/m}^2 \text{ sec Pa}$, which was 1/90 of that for direct Al coating on the polystyrene shell surface by electron beam vapor deposition. This permeability was almost the same as that obtained by the vapor deposition method. These success in suppressing tritium leakage enables us to fire polystyrene shells with gaseous DT at room temperature.

The amount of tritium in the shell is non-destructively characterized by counting x-rays from high Z material stimulated by β -rays from tritium decays. Figure 8 shows relation between the DT gas pressure and the count rate of the bremsstrahlung x-rays of Al. The x-ray counts were measured with a fluorescent x-ray analyzer and the DT gas pressure was measured with destructive method. The count

rate was proportional to the product of the fill pressure and the surface area. The fill pressure P_{DT} (MPa) could be experimentally given by ;

$$P_{DT} = 2.25 \times C / S$$

where C is x ray counts in 1/s and S is shell surface area in mm^2 .

Scattering of data was mainly caused by ambiguous thickness of the Al layer that was monitored just on a planner substrate.

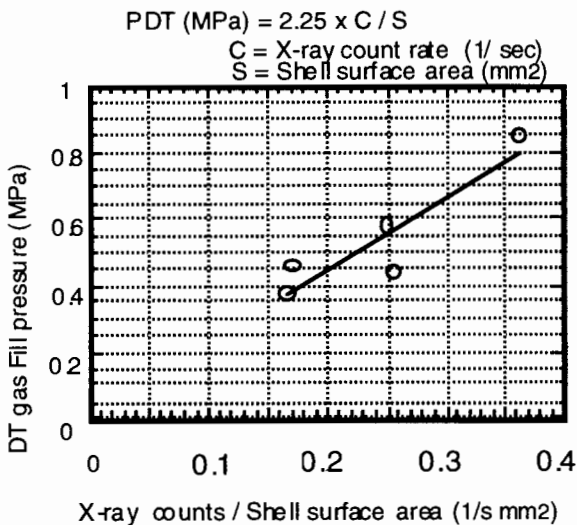


Fig. 8 Dependence of DT gas fill pressure measured by destructive method on x-ray count rate.

2.5 Fabrication of uniform solid DT layer

2.5.1 Principles to make uniform solid DT layer.

Cryogenic targets are required to demonstrate thermonuclear ignition and burn. Possibilities for making the cryogenic target are β -layering[7], IR layering[8], plasma layering[9] and foam method[10]. Among these techniques, the β -layering is mainly studied because it can redistribute a nonuniform, solid, deuterium-tritium (DT) layer into a uniform one by simply keeping the target in an isothermal environment. Because of the self heating of tritium decays, thicker layers of solid DT fuel tend to have warmer inner surfaces than thinner ones do. Due to the temperature dependent vapor pressure of the sold DT, a net sublimation of material from the thicker section to the thinner one takes place with an exponential rate constant as seen in Fig. 9. The temperature gradient in the solid also forces bubbles and cracks toward

the warmer region through the sublimation and deposition cycle.

It is known that the time constant for the redistribution is 28 min for fresh DT fuel without helium 3. In other words, it takes one hour to obtain a solid DT layer with thickness uniformity >99%. This time for stabilization is sufficient for current experiments. The long time for redistribution, however, results in large tritium inventory in a pellet factory of a future laser fusion plant. In addition this technique can not eliminate cracks and bubbles in the solid DT of which thickness is close to the average because the temperature gradient near the surface is almost zero, resulting no migration of voids.

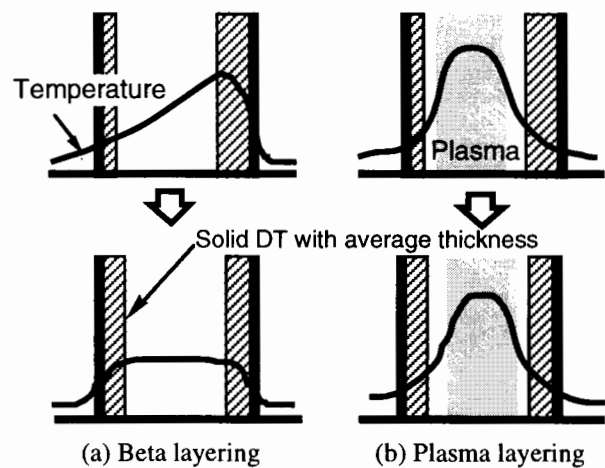


Fig. 9 Principle of beta layering (a) and plasma layering (b).

We have proposed the plasma layering technique to reduce the time to produce the uniform solid fuel layer. This technique can be used for smoothing both a solid deuterium and a deuterium-tritium fuel layer inside a plastic capsule. This technique can reduce the burden of handling radioactive tritium.

When a cryogenic target located in an isothermal environment has a uniform heat source at the center that can uniformly heat the inner surface of the solid fuel layer, the inner surface temperature of thicker parts becomes higher than that of thinner parts. This temperature difference enhances sublimation of the solid hydrogen in thicker parts and condensation of the vapor on thinner parts. As a result, the nonuniform solid fuel layer becomes uniform analogous to that seen in the β -laying method.

2.5.2 Issues

The principle of the plasma layering has already been demonstrated using Kr gas cooled by liquid nitrogen instead of deuterium with liquid helium coolant. However, the following issues remain to be solved.

- 1) Ultimate uniformity achievable under a one dimensional external electric field.

In current plasma layering technique, the target is located in a microwave resonator to initiate a glow discharge in the void of the target. Since the discharge is one dimensional, the heating uniformity strongly depends on the gas density in the void as shown in Fig. 10. Some technique is required to keep the discharge in an abnormal glow discharge region.

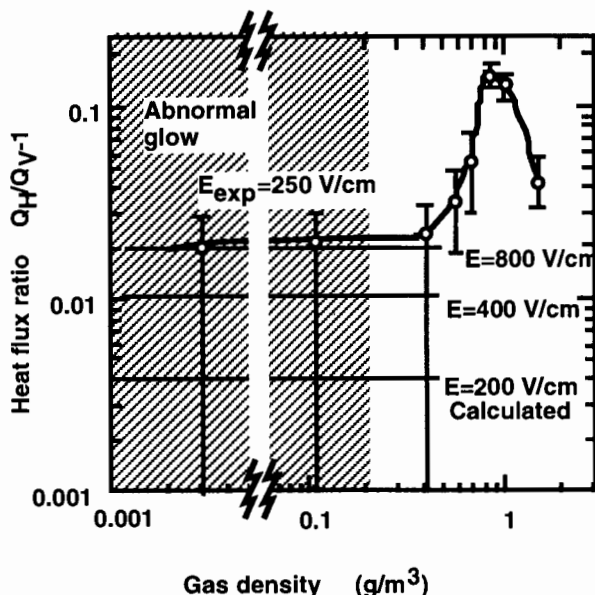


Fig. 10 Heating uniformity of plasma on inner surface of solid hydrogen layer.

- 2) Time constant for the redistribution.

The time constant is theoretically estimated to be primarily determined by the cooling rate of the target. An actual time constant in the system has to be studied.

- 3) Peeling-off of the solid deuterium due to large temperature gradient at the inner surface induced by plasma heating.

The discharge under a constant intensity of microwave field is expected to be intermittent, which may cause heat shock in the ice.

- 4) Cracks and deformation in the solid deuterium while heating the target from 11K for plasma layering to 17K for implosion.

The plasma layering will be carried out at 11K and laser irradiation of the cryogenic

target will be down at 17K. Since the hydrogen ice has a large volume expansion rate, cracks may occur during the heating process.

2.5.3 Experiment and result

We measured the time constant for the redistribution using a 6 mm diameter quartz shell with a gas feeder. The shell is housed at the center of a 10 K microwave cavity. After an appropriate amount of deuterium is frozen in the shell, 2.45 GHz microwave pulses are fed into the cavity through a coaxial cable. The power of the microwave in duty is 50 W. The pulse width and the repetition rate are varied from 0.1 to 100 ms and 500 to 10 pulses per second, respectively. Visible emission from the glow discharge is detected by a photomultiplier which shows the characteristics of the discharge.

When the pulse width of the microwave is longer than 0.2 ms, the discharge time has almost no dependence on pulse width of microwave. The longest discharge time is 0.2 ms which can be explained by a simple model in which dissipated power is used to heat the sublimated gas and an inner skin layer of solid deuterium. The discharge is automatically quenched by a rapid increase in vapor density in the void. In this case, the uniformity of the ice layer is insufficient because the glow discharge goes into a normal glow discharge region where the heating uniformity is low as shown in Fig. 10.

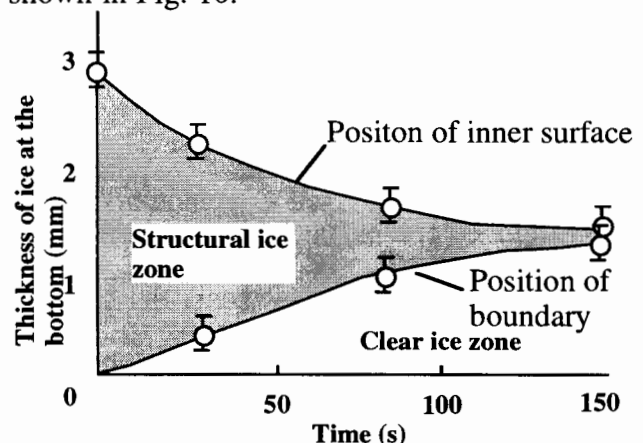


Fig. 11 Temporal change in thickness of solid deuterium.

When the width of the microwave pulse is less than half of the maximum discharge time, solid deuterium at the bottom of the shell is successfully redistributed to the upper region of the shell. The temporal change in thickness of the ice layer at the bottom of the shell is shown in Fig. 11. The thickness approaches

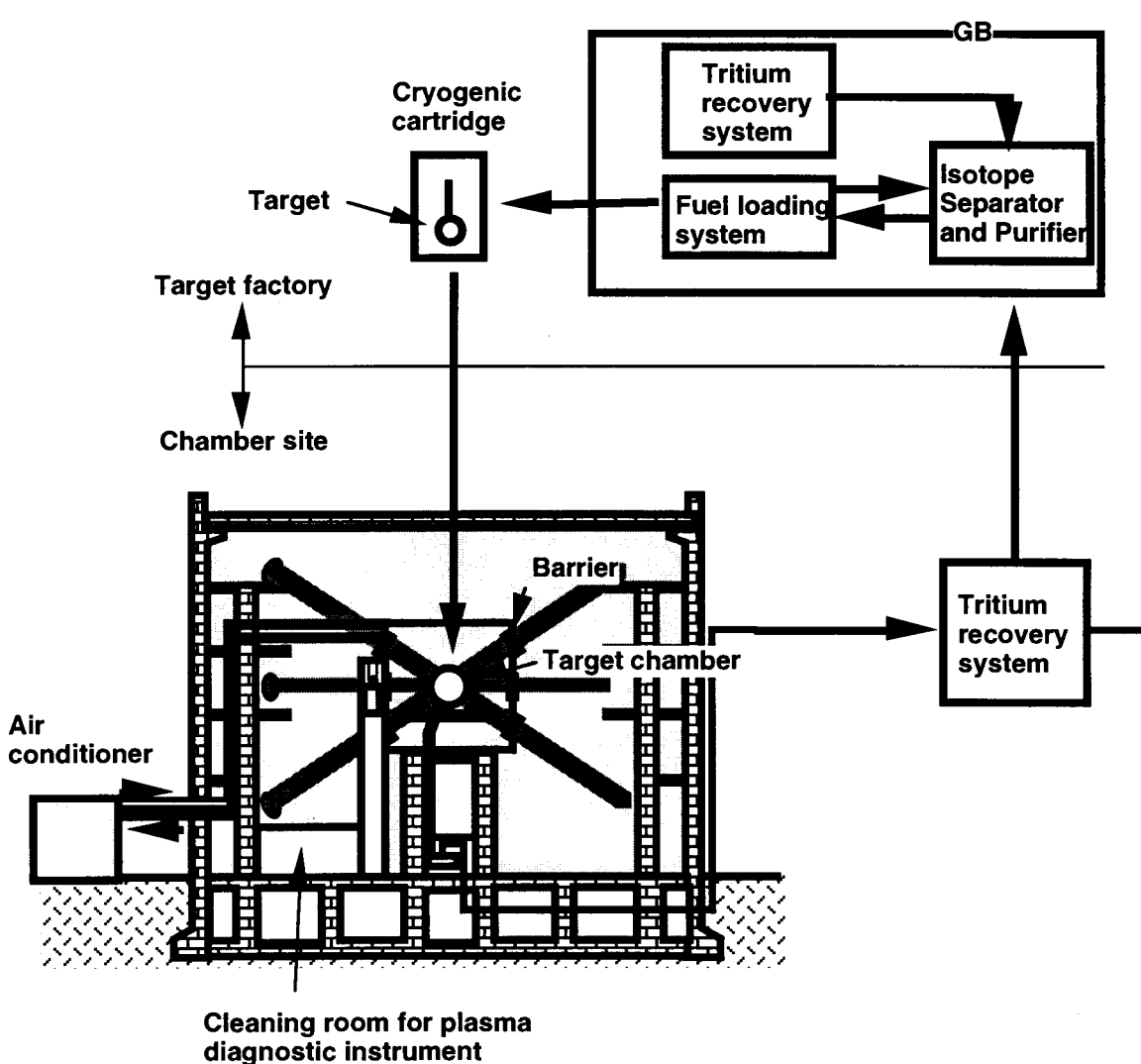


Fig 12 Proposed tritium facility around the target chamber

exponentially to its ultimate thickness and a clear ice zone appears from the bottom which is initially opaque due to cracks. The clear ice zone expands to the inner surface. From this exponential change, we can estimate that the time constant for the redistribution by plasma layering is 80 sec, that is 1/10 of the beta-laying method.

During discharges, no cracks larger than 10 μm were observed in the ice layer. After the redistribution is over, the shell is heated to the designated temperature. When the heating rate is less than 0.5 K/min, no crack is optically observed.

2.5.4 Summary

Smoothing of a solid deuterium layer by plasma layering is demonstrated for the first time and the time constant for the redistribution

was 80 sec. Experimental results indicate that the plasma layering can redistribute a nonuniform solid layer into a uniform one much faster than the β -laying, and can also eliminate cracks inside the ice layer. The shorter redistribution time is beneficial in reducing the tritium inventory in a future laser fusion reactor.

3. Issues for the next step

The next step of our experimental campaign is to demonstrate the ignition using a precision power-controlled laser. Plastic shells of which diameter, wall thickness and fill pressure at room temperature are 600 μm , 8 μm and 10 to 20 MPa, respectively, will be used in the campaign. The amount of tritium in the shell is about 37 GBq. The tritium facilities around the

target chamber will be improved as shown in Fig. 12. The target chamber will be housed in a sealed box equipped with a one-way air conditioner and the boundary of the hot zone is settled at the surface of the box. To remove tritium contamination on the plasma diagnostic equipment, a decontamination room will be located around the chamber.

To prevent an explosion, the temperature of the shell must be kept at less than 25 K during transportation of the shell from the fill station to the target chamber. Development of a compact portable cartridge with a reusable seal for high pressure tritium gas is necessary for this campaign. The uniform solid fuel layer will be produced by β -layering technique. Influence of isotope separation that takes place during the redistribution of solid DT through the sublimation-deposition cycle must be examined. Although this problem is avoided with the use of DT molecules, the current fuel is a mixture of D_2 , DT and T_2 . Light D_2 moves faster than heavy T_2 resulting rich in D at the deposited area and rich in T at the etched area. This isotope separation is from a nuclear DT reaction point of view but very for nonuniform ρR where ρ is the density of the solid fuel and R is the radius. The nonuniformity may cause Rayleigh-Taylor instability and degrade the implosion performance.

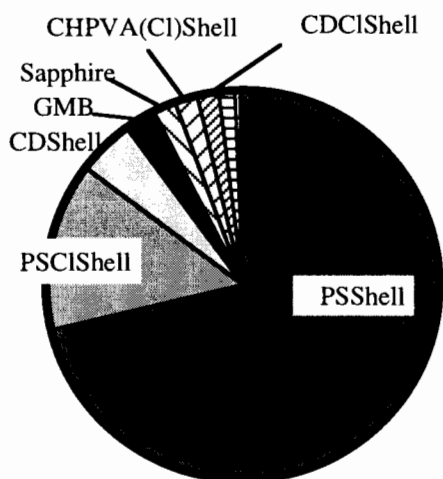


Fig. 13 Variety of targets fabricated in 1995.
 PSShell=polystyrene shell,
 PSCShell=polystyrene shell with Cl dopant,
 CHPVShell=above with a gas barrier made with polyvinyl alcohol,
 CDShell= deuterated polystyrene shell,
 GMB=glass microballoon

Behavior of tritium in the target chamber must be studied extensively to determine the handling of plasma diagnostic equipments around the chamber. In 1995, 1000 targets were fired in the target chamber. The variety of targets is shown in Fig. 13. The inner surface of the target chamber is covered with a thin layer of target debris which contains Au, B, C, Cl, Cu, F, Pb, Si and so on. Reaction of tritium under the existence of high energy photons is also one of our interests.

References

- [1] C. Yamanaka, S. Nakai, T. Yamanaka, Y. Izawa, Y. Kato, K. Mima, K. Nishihara, T. Mochizuki, M. Yamanaka, M. Nakatsuka, T. Sasaki, T. Yabe, K. Yoshida, H. Azechi, H. Nishimura, T. Norimatsu, S. Ido, N. Miyanaga, S. Sakabe, T. Jitsuno, M. Takagi, Nuclear Fusion, 27 (1987) 19.
- [2] H. Azechi, T. Jitsuno, T. Kanabe, M. Kata yama, K. Mima, N. Miyanaga, M. Nakai, S. Nakai, H. Nakaishi, M. Nakatsuka, A. Nishiguchi, P.A. Norrays, Y. Setsuhara, M. Tkagi, M. Yamanaka, C. Yamanaka, Laser and Part. Beams Vol. 9, No. 2, (1991) 193.
- [3] M. Takagi, T. Norimatsu, Y. Kato, and S. Nakai, Fusion Technology 14 (1988) 845.
- [4] M. Takagi, T. Norimatsu, T. Yamanaka, and S. Nakai, J. Vac. Sci. Technol., A10(1) (1992) 239.
- [5] M. Takagi, T. Norimatsu, T. Yamanaka, and S. Nakai, J. Vac. Sci. Technol., A9(4) (1991) 2145.
- [6] M. D. Wittman et al., Presented at 39th Nat. Sym. American Vac. Soc., Nov. 9-13, 1992, Chicago, USA.
- [7] A. J. Martin, R. J. Simms / R. B. Jacobs, J. Vac. Sci. Technol., A6(3) (1988) 1885.
- [8] Bittner, D., et al., Presented at 42nd Nat. Sym. America Vacuum Society, Oct. 16-20 (1995), Minneapolis, USA.
- [9] C. M. Chen, T. Norimatsu, Y. Izawa, and S. Nakai, J. Vac. Sci. Technol., A13(6), (1995) 2908.
- [10] T. Norimatsu, H. Ito, C. Chen, Y. Yasumoto, M. Takagi, K. A. Tanaka, T. Yamanaka and S. Nakai, Rev. Sci. Instrum., 63 (1992) 3378.