INERTIAL CONFINEMENT FUSION REACTOR DRY WALL STUDY

**FINAL REPORT FOR THE PERIOD
13 AUGUST 1981 - 31 MARCH 1983**

**Eugene W. Sucov
April 1983**

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**WESTINGHOUSE ADVANCED ENERGY SYSTEMS DIVISION
P.O. Box 158
Madison, PA 15663**

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EUGENE W. SUCOV

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1.0 ABSTRACT

The Westinghouse ICF Dry Wall Study was undertaken 1) to explore the practical implications of using a Ta coating to protect the steel first wall of an ICF reactor against the power pulses from the explosions of a pellet containing Ta as the heavy element (a concept which had emerged from a previous study to design an ICF Power Plant) and 2) to determine if a feasible design for improved safety and lower cost in a blanket could be developed using solid lithium compound in place of liquid lithium as the tritium breeder.

Three coating techniques were examined; plasma spray, chemical vapor deposition and explosive bonding. Plasma spray was chosen because it permits in-situ application, allows coating of irregularly shaped surfaces, can be scaled up to commercial dimensions and can be used to deposit other high temperature refractory materials. A thermal cycle test was designed to investigate the mechanical strength of the plasma sprayed Ta coating when cycled slowly ($\Delta T \sim 20^\circ\text{C}/\text{min}$) between room temperature and 500°C . This cycle produced thermal stresses in excess of those expected during the startup and shutdown of the ICF power plant after cycling. The samples were subjected to measurements of density, tensile and shear strength and evaluated with scanning electron microscopy (SEM) and x-ray diffraction (XRD). Tensile and shear strength of cycled samples was as good as that of uncycled samples. However, the plane of failure in the tests was always within the plasma sprayed layer, indicating that the bond was stronger than the Ta coating itself. SEM and XRD evaluations suggest the presence of a compound of tantalum at the fracture layer.

An evaporation code and a sputtering code which were developed at LANL, were used to calculate the loss rate of Ta due to these processes after each pellet explosion. While the surface temperature reaches to within 400°C of the melting temperature, it is not at these high temperatures long enough to generate significant losses by evaporation. Sputtering of Ta is substantial; however, it redeposits within 50 ms and therefore does not affect subsequent shots. The CHART D code calculates significant spallation of the front

surface due to absorption of the soft x-rays. By modifying the pellet design, the amount of spallation can be reduced to the point where excess T deposited by the standard pellet debris can be removed by spallation from special pellets.

A simulation experiment to verify the CHART D calculations was investigated. Sources of pulsed x-rays and ions to simulate the debris from each pellet explosion were identified. It was proposed to measure the instantaneous temperature of the surface with a two-color pyrometer focused on a very fast response time vidicon tube whose response would be stored in video tape for subsequent analysis. Stresses at the surface and at the back of the sample could be measured with holographic interferometry.

The fluidized bed concept continuously removes neutron generated heat and tritium from a blanket composed of solid lithium compounds. Helium gas at low pressure passes through and carries particles up through 32 meter high cylinders which are butted together to form the reactor wall. The design point value of 120 cm for the diameter of cylinders leads to a tritium breeding ratio of 1.2 which is more than adequate. By varying the flow velocity, the dwell time of the particles can be controlled to match either heat removal or tritium removal constraints.

The CANDID code was developed to permit evaluation of candidate metals for coating the steel based on criteria such as surface and bulk temperature rise, thermal stress in the creating layer and evaporation rate. Material properties were stored in the memory and were called upon to calculate evaluation algorithms. Of twenty original candidates, six remain: Re, Ir, Mo, Cr, W, Ta and Nb. Further evaluation would include parameters such as cost, manufacturability, radioactive decay rate, etc.

2.0 INTRODUCTION

The previous contract, DE-AC08-79-DP40086, produced a novel reaction chamber design in which the steel structural wall is protected against the destructive x-ray and ion pulses resulting from the pellet explosions by a thin coating of tantalum. The present contract, DE-AC08-81-DP40146, is mainly devoted to developing a more thorough and complete understanding of the limits and possibilities of the tantalum "dry wall" protection concept. In addition, a small effort was devoted to quantitatively evaluating various candidate metals to discover whether Ta was indeed the best choice for protecting the structural wall. Finally, a significant portion of the contract effort was devoted to the design of a blanket which uses solid lithium compounds instead of liquid lithium as the tritium breeding material. The solid lithium compounds are significantly less chemically reactive than the liquid lithium and therefore provide an increased measure of safety against adverse environmental impact in the event of an accident. Because of this feature, it is likely that the cost of the plant will be reduced.

The original contract terminated on August 12, 1982; however, the contract was extended to March 31, 1983 at no cost to the government.

The work to be done under this contract was divided into 7 tasks, as follows:

1. Selection of Metal for Pellet and First Wall Coating.

This task required that Ta and other metals be examined for their potential usefulness to protect the first wall. Specifically, a) a decision model was generated for selecting a metal and put into digital computer form, b) upgraded information on metal properties was collected for insertion into the computer model, c) candidate metals were rank ordered and one was selected for further study. The description of work done in Task 1 is found in Section 4.0.

2. Study Response of Ta coating to Pellet Yield (X-ray and Ion) Irradiation.

Specifically this task required that we a) calculate temperature and stress transients resulting from LANL provided pellet yield spectra from a Ta containing pellet, b) calculate evaporation and sputtering losses under the above conditions, c) explore possibility of equilibrating Ta deposition rate with Ta loss rate by adjusting pellet yield and pellet output spectra. This work is reported in Sections 5.2, 5.3 and 5.4.

3. Define Physical and Chemical Reactions in the Ta Coating.

This task contained two parts: a) explore chemical reactions taking place in Ta coating as debris ions strike it; determine effect of new compounds on ability of Ta coating to protect the steel substrate and b) explore consequences of collisions of debris ions and evaporated Ta with residual cavity gas. These tasks are described in Section 5.1.

4. Evaluate Manufacturability of the Reaction Chamber Wall.

The results of this task will be the selection/identification of a recommended process for manufacturing the Ta steel wall. The plasma spray method was chosen and is described in Section 6.0.

5. Examine Low Frequency Transient Effects at the Ta/Steel Interface.

Low frequency thermal stress is generated because of the differences in thermal expansion between the HT9 tubular wall and the Ta coating when the wall is heated from room to normal operating temperature (approximately 500°C). An experiment was performed to verify the integrity of the Ta bond to the steel under the startup-shutdown temperature cycle. Metallurgical inspection and analysis of the interface were made. This work is documented in Section 7.0.

6. Scope Experiment to Simulate Transient Effects in the Ta Coating.

This task initiated the design of an experiment which will simulate the transient conditions at the Ta coating due to x-ray and debris ions. Section 8.0 describes this work.

7. Evaluate Solid Lithium Blanket Concept.

A fluidized bed concept was selected for further examination because it had clear advantages over static bed concepts for removal of heat and tritium. A low gas pressure design was identified in which the heat and tritium are removed from the blanket by removing the particles themselves.

This effort is described in Section 9.0.

3.0 EXECUTIVE SUMMARY

In this study three major tasks were performed 1), the tantalum "dry wall" concept was more thoroughly investigated, 2), alternate metals to Ta were evaluated and ranked, and finally, 3) a blanket concept utilizing solid lithium compounds as the tritium breeder was evaluated.

Three techniques were examined for applying the 1mm Ta coating to a steel substrate: plasma spray, chemical vapor deposition and explosive bonding. Advantages of the plasma spray method, compared to the two alternative techniques, include:

- o Allows thick deposits on large, irregularly shaped surfaces.
- o Permits in-situ coating or repairs.
- o Can be scaled up to the referenced ICF structural configuration and dimensions.
- o Experience with a wide variety of coatings (including tantalum) and substrates shows good thermal fatigue and shock resistance.
- o Can be used to deposit other high temperature refractory materials (metals or ceramics).

The spraying was performed with the gun held ~3 inches from the substrate (stainless steel) and the spray was swept both parallel and perpendicular to the axis of the cylinder. Spraying was continued for several minutes at a time and stopped when the substrate began to heat up. There was enough fugitive oxygen in the present system to cause some oxidation of the sprayed tantalum which was noticeable when the substrate heated up to a few hundred degrees centigrade. After the substrate had cooled again, the process was repeated until the desired thickness was built up. Typically 5-10 mills were added per spraying using the coarser powder (#1) and 2-4 mills using the finer (#2).

The surface of the plasma sprayed tantalum has an overall matte finish. On close inspection, small, shiny metallic areas are evident. These are the surfaces of the last droplets to splat against the sprayed surface. Other miscellaneous observations indicate that the plasma sprayed layer is both hard and brittle. Striking the layer on an edge will not indent the layer but cause pieces to chip off. Significantly, the breaks always occur through the bulk of the layer and never at the layer-stainless interface. A steel file will mark the surface of the layer with difficulty and when drilled from the stainless through to the plasma sprayed layer, the tantalum tends to spall off around the hole, but again not breaking at the tantalum-stainless interface.

A thermal cycle test was designed to demonstrate the mechanical stability of the plasma sprayed tantalum coating on HT-9 stainless steel when cycled slowly ($\Delta T \leq 20^\circ\text{C}/\text{min}$) between room temperature and 500°C . This cycle produced thermal stresses in excess of those expected during the startup and shutdown of the ICF power plant.

Two 12 inch pieces were each cut into four 3 inch sections, then drilled and restraining bolts installed so that each half cylinder would not be able to warp during subsequent thermal cycling. They were encapsulated in a pyrex envelope under high purity argon gas ($\sim 2/3$ atm at room temperature) to prevent oxidation of the tantalum at elevated temperature in the furnace. Tubulations at the top and bottom were used to flush the samples with high purity argon during encapsulation.

The encapsulated test pieces were loaded onto an elevator platform which was periodically lowered into and raised out of the hot zone of the furnace below. Stainless steel plates acted as baffles to help control convection currents around the test pieces. The temperature was monitored by a chromel-alumel thermocouple inserted from the top which indicated the temperature at the mid-point of the samples. A dummy sample (two pieces of stainless steel in an open glass cylinder) was installed to replace actual test samples as they were removed. This was done to maintain a similar thermal regime throughout the test.

In the "up" position the samples were cooled further by a small fan blowing on them. The average high and low temperatures were $500 \pm 15^{\circ}\text{C}$ and $35 \pm 5^{\circ}\text{C}$. In total there were four sets of two samples; each set consisted of one sample sprayed with #1 (-100 +200) Ta and one sample sprayed with #2 (-200 +325) Ta. The four sets experienced 0, 20, 50, and 100 cycles respectively. This coating was not degraded by thermal cycling to 500°C under an argon atmosphere and had mechanical strength levels more than adequate to support itself reliably.

Our evaluation of the coating both by visual and instrumental analysis suggested that other phases were present. The fact that only tantalum was detected by the scanning electron microscope indicates that only elements lighter than sodium could be present in addition to tantalum, a fact consistent with the X-ray diffraction analyses results, which indicated the presence of an oxide and a nitride. The other element probably present is hydrogen which is readily soluble in tantalum at red heat and could have been picked up from the arc gas during plasma spraying. Since tantalum forms a variety of nitrides and oxynitrides in addition to oxides and hydrides, including limited solid solutions with these elements, it is not possible to sort out all the possibilities with the limited analytical data available. However, all of the data are consistent with these types of compounds producing the observed heterogeneity.

The brittleness of the tantalum coating mentioned earlier is certainly the result of chemical contamination by oxygen, nitrogen and hydrogen. Since one of the attractive properties of tantalum metal as a first surface coating is its malleability, an effort should be made to reduce the level of gas (O, N, H) contamination so that a malleable coating is produced. Tantalum is quite malleable if its oxygen content is below 400-600 ppm. The present coating has about twice that much oxygen in the starting material and an unknown amount from the plasma spraying procedure. The final amounts of hydrogen and nitrogen are also unknown. The potential sources of contamination are known but their relative importance is not known at this time.

The reactions between tantalum and the debris ions from the pellet explosion were examined in terms of the depth of penetration of implantation of the

debris ions and the rate of implantation. Descriptions of unclassified pellets were used to estimate the number and species of debris ions, and solubility and diffusivity data were used to estimate the nature of the chemical reactions of these ions with Ta. It was found that:

- A. The accumulation of H, D, and T, and the formation of hydrogen bubbles on the first wall will be negligible.
- B. The ion implantation of the heavier debris ions will be approximately 0.5 μm at most.
- C. The carbon and silicon will form stable, non-volatile carbides and silicides via reactions with the Ta substrate and with implanted Ta.
- D. It is likely that the implantation of the debris will cause high surface stresses on the wall, with subsequent surface damage and spallation. But this must be investigated experimentally to determine the magnitude of the effect.
- E. In the absence of excessive spallation the first wall should increase, or maintain, its thickness via the accumulation of the debris.

The CHART D calculations which had been used to confirm the viability of the dry wall concept had been based on classified data for a pellet whose heavy metal was not tantalum. It became necessary, therefore, to redo the calculations with yield data from a pellet using Ta as the heavy metal. This classified data was generated at Los Alamos and supplied to us. The CHART D calculations identified temperature and stress conditions at the surface and within the Ta coating as well as failure of the surface by spallation.

There were two main objectives to these calculations. In the first objective, the pellet parameters were adjusted until the corresponding soft X-ray deposition profile in the first wall coating did not result in any evaporation or spalling. In second objective, a deposition profile was used which caused the spallation in a controlled manner of several layers of tantalum. In this

case, the pellet will be used to reduce the build up to Ta on the first wall by the spalling and subsequent removal from the pellet chamber of the spalled material.

A scoping study investigated the design of a facility which could simulate the x-ray and ion pulses characteristic of a fusion reactor. Three sources of x-rays were considered, a) characteristic, b) bremsstrahlung and c) transition. Bremsstrahlung is relatively strong at short wave lengths while transition is strong at long wavelengths; the two forms complement each other. However, it was found that special electron guns were needed in order to be able to generate the required intensities within pulses at least 200 ns in duration. An alternative source of very intense, short pulse irradiation was found to be either the chemical laser or the CO₂ laser. Two kinds of ion sources were considered, 1) field ionization and 2) large area sparks. The latter seemed to satisfy the experimental requirements.

Temperature measurements by two color optical pyrometry are accurate to about 1% and were found to be capable of following the rapid transients when used with a photomultiplier coupled to a vidicon detector. Measurements of surface deformation are conveniently done by using the techniques of pulsed holographic interferometry using a Q-switched ruby laser.

Using the CANDID code, 20 pure metals were investigated for potential application as first wall coatings for ICF reactor chambers. The data base of material properties included 19 physical parameters for each of the 20 candidate metals. Based on a number of these parameters as well as on several key ICF reactor parameters, six figures-of-merit (FOM) were calculated using CANDID for each candidate. By taking weighted sums of the six FOM values, overall FOM values were obtained. The six FOM calculations are based on mechanical, thermal, and vacuum vaporization engineering constraints. The 20 pure metal candidates investigated were aluminum, silver, gold, beryllium, chromium, copper, iron, iridium, molybdenum, niobium, nickel, platinum, rhenium, silicon, tantalum, thorium, titanium, vanadium, tungsten, and zirconium.

The candidate materials recommended for further consideration as first wall coatings are

- A. Rhenium (Re)
- B. Iridium (Ir)
- C. Molybdenum (Mo)
- D. Chromium (Cr)
- E. Tungsten (W)
- F. Tantalum (Ta)
- G. Niobium (Nb)

where the list is in rank order based on FOM values. Tantalum is the material previously selected for the ICF dry wall design.

One of the contract objectives, prompted by safety and cost considerations, is design of a blanket that uses solid lithium compounds as tritium breeding materials. Three blanket design options were considered initially: 1) liquids other than lithium; 2) stationary solids; and 3) circulating solids. Lead lithium eutectics, specifically $\text{Li}_{17}\text{Pb}_{83}$, were candidates for the liquid blanket, but have several disadvantages: low thermal conductivity, low lithium content, corrosive behavior similar to that of liquid lead, high density, and poorly-known thermal and chemical properties. A static solid blanket requires a relatively complex heat removal system, since helium or water must be circulated throughout the blanket, as well as a system for in-situ removal of tritium. The success of the latter depends on tritium release characteristics, which are uncertain and may be significantly altered by effects of irradiation and cyclic stresses on the microstructure. The third design concept, which employs moving solids, includes both conventional packed beds and fluidized beds. Tritium removal from the former has problems similar to those in the static beds, but the latter appears to have a number of advantages, not the least of which includes potential for recycling the bed material if effects due to radiation damage or tritium accumulation are unacceptable.

Mechanical design of two concepts for fluidizing the bed and carrying off excess heat and tritium were performed. One concept utilizes a high pressure

stream of helium to remove the heat generated in the particles by neutron absorption; tritium did not diffuse fast enough into the gas stream so it was removed by moving the particles with the gas stream into an external collector. The other concept uses a low pressure helium stream to fluidize the bed but removes the heat and the tritium by blowing the particles out of the blanket and into an external collector. In the high pressure approach, helium gas at 800 psi not only fluidizes the particles but it also is used as the sole method of heat removal. In the low pressure approach helium gas at 50 psi fluidizes the particles but carries away only 1% of the absorbed neutron energy. The remainder of the energy is carried out of the blanket by bleeding off the hot particles to an auxiliary fluidized bed heat exchanger. The low pressure design was significantly simpler to construct and promised improved tritium breeding so it was chosen as the reference design.

The low pressure fluidized bed blanket which forms the cylindrical wall of the chamber is made up of thirty-six tall steel tanks which run the full thirty-two meter height of the chamber. Each of these tanks forms the containment for an individual fluidized bed of sintered lithium oxide pellets (which have a maximum diameter of one millimeter) suspended in helium gas at 50 psi. The blanket has an average radial thickness of two meters. Neutrons give up their energy to the suspended particles which are in turn continuously withdrawn at 475°C from the top of each tank to carry the heat to the fluidized bed heat exchangers which serve as steam generators. The cooled particles at 300°C are then returned to the bottom of the fluidized bed tank by a screw feeder. About 1% of the neutron energy is carried away by the fluidizing gas to a separate heat exchanger. Horizontal fluidized beds two meters deep, contained in tanks two meters wide, form the top and bottom of the cylindrical reaction chamber. Again the heat is removed from the fluidized beds by withdrawing the hot particles. The 300°C particles are fed into one end of the horizontal fluidized bed and flow by gravity like a liquid to be spilled out at the far end at 475°C. The particles are transported by fluidized bed piping to the fluidized bed heat exchangers. A small portion of the circulating particle stream will be withdrawn continuously for heating to a high temperature to drive off the tritium. These particles will be

pulverized, mixed with the fines separated from the fluidizing gas and resintered into particles of the size required for proper operation of the fluidized beds.

Fluidized beds have many advantages over static beds for a neutron absorbing blanket.

- A. The particles of the lithium compound which absorb the neutrons and become hot are always in intimate contact with the helium coolant. In contrast to the static bed design, the fluidized bed does away with the hundreds of thousands of tiny tubes which would have to be placed in solid but porous blocks of lithium compound in order to provide a low thermal drop between the solid and the coolant. Also, the very complex manifolding required to bring the coolant to all these small coolant tubes is eliminated. The thousands of welded tubes needed in the static bed design give a very high probability for leaks. In the tank design for the fluidized beds, there are no delicate welds and the number of welds is small. Leaks if they occur would be outside the plasma vacuum space.
- B. A fluidized bed is characteristically isothermal because the agitated particles cause the bed to have a very high thermal conductivity. Thus, even though the neutron absorption is highest on the tank side facing the exploding pellet, the temperature is nearly uniform throughout the bed. This permits a higher average blanket temperature for a given inner tank wall temperature.
- C. The particles of the lithium compound are bled off continuously to permit the removal of the tritium. This greatly reduces the tritium inventory.
- D. Fluidized beds are a commonly used technology in the chemical processing industry. They have formed the basis for the catalytic cracking of gasoline since the early 1940s. Fluidized bed design practices are well established and the cost of developing a fluidized blanket will be low.

The solid angle weighted global tritium breeding ratio (TBR) is 1.27, exceeding the design goal of 1.2 by a substantial margin. One reason for this improved breeding performance, compared to the high pressure fluidized bed design, is the lower structural volume fraction. This results in a parasitic absorption rate of less than 10%.

Analysis of the cumulative tritium breeding ratio as a function of bed thickness shows that the TBR rises rapidly at first, slowing down above about 80 cm in bed thickness, and then approaching the full bed thickness (200 cm) value of 1.28 in a near asymptotic fashion. The target value of 1.20 occurs at a thickness of approximately 120 cm. Thus, on the basis of TBR requirements, it appears that a considerable reduction in fluidized bed thickness is possible. However, an additional neutronics requirement is adequate energy deposition in the blanket region. Analysis of this problem showed that at a blanket thickness of 120 cm, which is adequate for tritium breeding, nearly 97% of the available neutron energy is deposited in the first wall and blanket. If the surface heat deposition of 1120 MW is included, 98% of the total energy is deposited. Moving the graphite reflector adjacent to this thinner blanket would raise this close to the 99% goal.

The average first wall neutron loading is 1.3 MW/m^2 , rising to a maximum value of 1.9 MW/m^2 at the surfaces nearest to the chamber center. Fluence lifetime under ICF reactor conditions for HT-9 is not well defined, but we can probably assume it to be close to that of titanium modified 316-SS (prime candidate alloy). Under low thermal stress steady state conditions this has been estimated to be 20 MWy/m^2 . These conditions are reasonably well satisfied in the present case. On this basis the expected first wall lifetime is therefore approximately 10 years.

4.0 SELECTION OF METAL FOR PELLET AND FIRST WALL

4.1 Introduction

The engineering solutions to the protection of the first solid surface of ICF reactor chambers include flowing liquid metals, thin liquid metal films, high density gases, magnetic fields, and thin metallic coatings, as well as various combinations of each of these schemes. Only the scheme using metallic coatings uses another solid material to protect the chamber structural wall. Such coatings must simultaneously satisfy mechanical, thermal, and vacuum vaporization engineering constraints during reactor operation, and these constraints seriously limit the number of materials to be considered. If these constraints are then coupled to considerations of manufacturability, coating technology development, and irradiation damage at elevated temperatures under pulsed loadings, the potential candidates for first wall coatings will more than likely be restricted to a mere handful of materials.

This section describes a brief study to initiate a selection process for establishing coating candidates. The study is limited to considering mechanical, thermal, and vacuum vaporization engineering constraints as they apply to pure metals. Considerations of cost, fabrication, radiation effects, and material compatibility were not included in this initial effort. The establishment of a set of algorithms for compilation of physical data and figures-of-merit was achieved. A code based on the algorithms was created and used to generate a ranking of candidates for first wall coatings. The algorithms and code are described in the next section. In Section 4.3 the results of the first study using this code are presented. Recommendations and a summary are given in Section 4.4.

4.2 Coating Candidate Selection and Ranking Method

A computer code was developed to select and rank materials. The code contains a data bank for physical properties of materials and a relatively simple tool for rank ordering these materials. The rank ordering is accomplished by a set of relatively simple algorithms to obtain numerical values for

figures-of-merit (FOM) which not only account for material properties, but also for features of the reactor cavity and the pulsed energy produced by the pellet explosions. The algorithms and code have been developed in a manner which permits additional materials, material properties, and/or FOM to be incorporated at a later time.

The code, which has been named CANDID (short for CANDIDATE DATA AND RANKING), has data for twenty pure metals. The data incorporated for each candidate consists of fourteen physical and mechanical properties which are either temperature independent, very weakly dependent on temperature, or have values only at room temperature (20-25°C). There are also five more physical and mechanical properties incorporated which are treated as temperature dependent and the FOM values are calculated accordingly. Tables 4-1, 4-2, 4-3, 4-4, and 4-5 are copies of the complete set of properties collected^(1,2) for this study and printed as output from CANDID.

The FOM calculations are based on estimating a number of first wall parameters related to the operation of the device and the coating material properties. These parameters are converted into a set of dimensionless variables ranging from zero to unity where the higher the value, the higher the potential merit of the material. This set is then combined into a single FOM per material by taking a weighted sum of these normalized values. The weights are established by selecting the relative importance of each individual term. Since there is no obvious means of setting weights, a number of weighting schemes were used (and are discussed at the end of this section).

The six independent FOM parameters selected for this study were based on the following parameters:

- A. The maximum temperature rise at the coating surface due to the energy pulse. This parameter indicates the design margin in maintaining the coating below melting conditions.

TABLE 4-1

PHYSICAL PROPERTIES OF COATING CANDIDATES ALUMINUM, SILVER, GOLD,
BERYLLIUM, CHROMIUM, COPPER, AND IRON

SYM, ATOMIC SYMBOL	AL	AG	AU	BE	CR	CU	FE
AM, ATOMIC MASS (U)	26.982	107.868	196.967	9.012	51.996	63.540	55.847
SC, CRYSTAL STRUCTURE (FCC,BCC,CPH)*	1.000	1.000	1.000	3.000	2.000	1.000	1.200
A, LATTICE CONSTANT (NM)	.40496	.40862	.40786	.22858	.28844	.36151	.28660
RHO, DENSITY (G/CM**3) AT 20-25 C	2.699	19.490	19.302	1.848	7.190	8.930	7.870
TM, TEMPERATURE AT MELTING (K)	933.60	1235.10	1337.60	1563.00	2148.00	1358.00	1811.00
SIG, TH. NEUTRON CROSS SECTION (B)	.700	0.000	0.000	.010	0.000	0.000	0.000
HVAP, LAT. HEAT OF VAPOR. (KJ/KG)	1.078E+04	2.630E+03	1.700E+00	0.	6.168E+03	4.729E+03	7.018E+03
SE, ENTROPY (J/KG**1 K**1)	1.050E+03	3.560E+02	0.	1.060E+02	4.600E+02	5.250E+02	4.900E+02
PV, VAPOR PRESSURE AT TM (TORR)	1.125E-06	3.333E-03	1.011E-04	4.777E-02	6.480E+00	5.029E-04	4.258E-02
ERATE, EVAPORATION (G CM**2 S**1)	1.104E-10	5.666E-05	2.240E-06	2.094E-04	5.820E-02	6.280E-06	4.316E-04
POIS, POISSON'S RATIO AT 20 C	.240	.380	.300	.048	.300	.350	.291
GPOD, SHEAR MODULUS AT 20 C (GPA)	25.00	26.00	0.00	137.00	78.00	47.00	81.00
ST, TENSILE STRENGTH (MPA)	45.0	125.0	103.0	290.0	200.0	205.0	300.0
HD, HARDNESS (HB, HRC, HRF, HV)	44.10	25.40	0.00	80.30	0.00	80.10	40.40
T, TEMPERATURE (K)	293.00	293.00	293.00	293.00	293.00	293.00	293.00
TCON, T. CONDUCTIVITY (W CM**1 K**1)	247.00	428.00	318.00	190.00	67.00	401.00	84.00
CP, SPECIFIC HEAT (J KG**1 K**1)	900.00	235.00	128.00	1886.00	460.00	386.00	430.00
ALPHA, EXPANSION (E-6M CM**1 K**1)	27.40	20.60	14.20	16.50	6.20	16.50	12.00
YPOD, TENSILE MODULUS (GPA)	62.00	71.00	78.00	288.00	210.00	128.00	190.00
YD, YIELD STRENGTH (MPA)	20.00	55.00	207.00	224.00	135.00	33.00	280.00
T, TEMPERATURE (K)	773.00	773.00	773.00	773.00	773.00	773.00	773.00
TCON, T. CONDUCTIVITY (W CM**1 K**1)	370.00	356.00	318.00	190.00	76.00	373.00	45.00
CP, SPECIFIC HEAT (J KG**1 K**1)	900.00	239.00	140.00	1886.00	460.00	420.00	645.00
ALPHA, EXPANSION (E-6M CM**1 K**1)	27.40	20.60	14.20	16.50	6.20	25.00	16.00
YPOD, TENSILE MODULUS (GPA)	62.00	71.00	78.00	288.00	289.00	128.00	208.00
YD, YIELD STRENGTH (MPA)	20.00	55.00	207.00	224.00	135.00	33.00	175.00

*1=FCC (Face centered cubic)
2=BCC (Body-centered cubic)
3=CPH (Close-packed hexagonal)

1.2 indicates a phase change from
FCC to BCC

TABLE 4-2

PHYSICAL PROPERTIES OF COATING CANDIDATES IRIIDIUM, MOLYBDENUM,
NIOBIUM, NICKEL, PLATINUM, RHENIUM, AND SILICON

SYM, ATOMIC SYMBOL	IR	MO	NB	NI	PT	RE	SI
AM, ATOMIC MASS (U)	192.22	95.94	92.906	58.710	195.084	186.207	28.086
SC, CRYSTAL STRUCTURE (FCC, BCC, CFH)*	1.000	2.000	2.000	1.000	3.000	1.000	1.000
A, LATTICE CONSTANT (NM)	.357	.357	.357	.357	.357	.357	.357
RD, DENSITY (G/CM**3) AT 20-25 C	22.65	10.22	8.57	8.902	21.45	21.74	2.33
TM, TEMPERATURE AT MELTING (K)	2720.00	2863.00	2741.00	1726.00	2042.00	3453.00	1687.00
SIG, TH. MELTING CROSS SECTION (B)	0.000	0.000	1.100	0.000	0.000	0.000	0.000
HVAF, LAT. HEAT OF VAPOR. (KJ/KG)	0.	5.12E+03	7.45E+03	0.	0.	0.	1.061E+04
SE, ENTROPY (J KG**1 K**1)	1.500E+02	3.000E+02	3.750E+02	5.150E+02	0.	2.250E+02	6.650E+02
VP, VAPOR PRESSURE AT TM (TORR)	6.509E-02	1.410E-02	1.38E-03	6.989E-03	2.707E-04	5.322E-02	2.052E-03
ERATE, EVAPORATION (G CM**2 S**1)	1.001E-03	1.485E-04	1.46E-05	7.441E-05	4.830E-06	7.134E-04	1.530E-05
PCIS, PCISSCAS FATIG AT 20 C	.260	.320	.380	.310	.300	.490	.300
GPOD, SHEAR MODULUS AT 20 C (GPA)	205.00	147.00	38.00	76.00	0.00	154.00	0.00
ST, TENSILE STRENGTH (MPA)	1170.0	775.0	275.0	217.0	207.0	1160.0	125.0
HD, HARDNESS (HRE, HK, HRC, HV)	22.40	100.40	80.40	64.40	0.00	270.40	0.00
T, TEMPERATURE (K)	293.00	293.00	293.00	293.00	293.00	293.00	293.00
TCON, T. CONDUCTIVITY (W M**1 K**1)	147.00	142.00	52.00	63.00	71.00	71.00	84.00
CP, SPECIFIC HEAT (J KG**1 K**1)	130.00	276.00	270.00	471.00	132.00	146.00	678.00
ALPHA, EXPANSION (E-6M M**1 K**1)	6.80	1.60	7.40	13.30	9.10	10.01	7.90
YPOD, TENSILE MODULUS (GPA)	517.00	320.00	103.00	207.00	172.00	460.00	113.00
YO, YIELD STRENGTH (MPA)	234.00	400.00	207.00	59.00	186.00	530.00	125.00
T, TEMPERATURE (K)	773.00	773.00	773.00	773.00	773.00	773.00	773.00
TCON, T. CONDUCTIVITY (W M**1 K**1)	147.00	122.00	63.00	63.00	71.00	71.00	84.00
CP, SPECIFIC HEAT (J KG**1 K**1)	130.00	276.00	280.00	471.00	132.00	146.00	678.00
ALPHA, EXPANSION (E-6M M**1 K**1)	6.80	3.40	8.20	13.30	9.10	10.01	6.80
YPOD, TENSILE MODULUS (GPA)	517.00	260.00	95.00	207.00	172.00	460.00	113.00
YO, YIELD STRENGTH (MPA)	234.00	125.00	130.00	59.00	186.00	530.00	125.00

*See footnote in Table 1

TABLE 4-3

PHYSICAL PROPERTIES OF COATING CANDIDATES TANTALUM, THORIUM,
TITANIUM, VANADIUM, TUNGSTEN, AND ZIRCONIUM

SYM, ATOMIC SYMBOL	TA	TH	TI	V	W	ZR
AP, ATOMIC MASS (U)	180.948	232.038	47.900	50.941	183.850	91.224
SC, CRYSTAL STRUCTURE (FCC,BCC,CFH)*	2.000	1.000	3.200	2.000	2.000	3.200
A, LATTICE CONSTANT (Å)	.33026	.50860	.29503	.35278	.31652	.35312
RHO, DENSITY (G/CM**3) AT 20-25 C	16.690	11.720	4.507	6.100	19.254	6.505
TM, TEMPERATURE AT MELTING (K)	3265.00	2028.00	1541.00	2173.00	3683.00	2125.00
SIG, TH. NEUTRON CROSS SECTION (B)	21.000	0.000	0.000	4.700	0.000	0.000
HVAP, LAT. HEAT OF VAPOR. (KJ/KG)	4.215E+03	0.	9.830E+03	0.	0.	6.520E+03
SE, ENTROPY (J KG**1 K**1)	2.200E+02	0.	6.320E+02	5.790E+02	1.820E+02	4.210E+02
PV, VAPOR PRESSURE AT TM (TORR)	5.567E-03	6.175E-05	6.050E-03	2.524E-02	3.726E-02	2.243E-05
ERATE, EVAPORATION (G CM**2 S**1)	1.299E-04	1.200E-06	5.486E-05	2.584E-04	4.805E-04	2.682E-07
FCIS, FRICTION FACTOR AT 20 C	.350	.270	.350	.360	.290	.350
GPCD, SHEAR MODULUS AT 20 C (GPA)	69.00	28.00	35.00	46.00	160.00	32.00
ST, TENSILE STRENGTH (MPA)	200.0	219.0	235.0	53.0	350.0	441.0
HC, HARDNESS (H, HK, HFB, HV)	110.40	185.40	72.10	72.10	100.40	0.00
T, TEMPERATURE (K)	293.00	293.00	293.00	293.00	293.00	293.00
TCON, T. CONDUCTIVITY (W M**1 K**1)	54.00	50.00	20.00	31.00	160.00	21.00
CP, SPECIFIC HEAT (J KG**1 K**1)	135.00	118.00	669.00	458.00	136.00	250.00
ALPHA, EXPANSION (E-6M M**1 K**1)	6.50	11.40	8.40	9.60	3.40	5.90
YMOD, TENSILE MODULUS (GPA)	186.00	72.00	127.00	124.00	410.00	59.00
YC, YIELD STRENGTH (MPA)	180.00	144.00	140.00	53.00	150.00	360.00
T, TEMPERATURE (K)	773.00	773.00	773.00	773.00	773.00	773.00
TCON, T. CONDUCTIVITY (W M**1 K**1)	66.00	50.00	20.00	31.00	130.00	22.00
CP, SPECIFIC HEAT (J KG**1 K**1)	153.00	156.00	669.00	458.00	143.00	290.00
ALPHA, EXPANSION (E-6M M**1 K**1)	6.80	12.60	8.40	9.60	4.80	5.90
YMOD, TENSILE MODULUS (GPA)	166.00	72.00	127.00	124.00	390.00	55.00
YC, YIELD STRENGTH (MPA)	50.00	144.00	140.00	53.00	125.00	360.00

*See footnote in Table 1

TABLE 4-4

TEMPERATURE DEPENDENT PROPERTIES FOR COATING CANDIDATES
MOLYBDENUM, NIOBIUM, AND TANTALUM

SPECIAL CASE FOR CANDIDATE MO AT 6 TEMPERATURES

T , TEMPERATURE (K)	273.00	523.00	773.00	1023.00	1273.00	1523.00
TCOA, T. CONDUCTIVITY (W M ⁻¹ K ⁻¹)	142.00	122.00	122.00	111.00	101.00	91.00
CP , SPECIFIC HEAT (J KG ⁻¹ K ⁻¹)	276.00	276.00	276.00	285.00	310.00	333.00
ALPHA, EXPANSION (E-6M M ⁻¹ K ⁻¹)	1.60	2.80	3.40	3.90	4.50	5.10
YPC, TENSILE MODULUS (GPA)	320.00	302.00	280.00	250.00	220.00	190.00
YC , YIELD STRENGTH (MPA)	400.00	150.00	125.00	100.00	85.00	50.00

SPECIAL CASE FOR CANDIDATE NB AT 6 TEMPERATURES

T , TEMPERATURE (K)	273.00	523.00	773.00	1023.00	1273.00	1523.00
TCOA, T. CONDUCTIVITY (W M ⁻¹ K ⁻¹)	52.00	58.00	63.00	68.00	74.00	79.00
CP , SPECIFIC HEAT (J KG ⁻¹ K ⁻¹)	270.00	275.00	280.00	300.00	315.00	330.00
ALPHA, EXPANSION (E-6M M ⁻¹ K ⁻¹)	7.40	7.70	8.20	8.60	9.00	9.40
YPC, TENSILE MODULUS (GPA)	103.00	100.00	95.00	90.00	85.00	80.00
YC , YIELD STRENGTH (MPA)	207.00	200.00	130.00	40.00	30.00	20.00

SPECIAL CASE FOR CANDIDATE TA AT 6 TEMPERATURES

T , TEMPERATURE (K)	273.00	773.00	1273.00	1773.00	2273.00	2773.00
TCOA, T. CONDUCTIVITY (W M ⁻¹ K ⁻¹)	54.00	66.00	78.00	80.00	82.00	85.00
CP , SPECIFIC HEAT (J KG ⁻¹ K ⁻¹)	139.00	153.00	164.00	174.00	186.00	198.00
ALPHA, EXPANSION (E-6M M ⁻¹ K ⁻¹)	6.40	6.70	7.00	7.30	7.70	8.10
YPC, TENSILE MODULUS (GPA)	186.00	168.00	150.00	131.00	113.00	94.00
YC , YIELD STRENGTH (MPA)	180.00	50.00	38.00	25.00	10.00	10.00

TABLE 4-5

TEMPERATURE DEPENDENT PROPERTIES FOR COATING CANDIDATES
TUNGSTEN, IRIIDIUM, AND IRON

SPECIAL CASE FOR CANDIDATE W AT 6 TEMPERATURES

T , TEMPERATURE (K)	273.00	773.00	1273.00	1773.00	2273.00	2773.00
TCON,T. CONDUCTIVITY (W M**-1 K**-1)	160.00	130.00	120.00	105.00	95.00	50.00
CP , SPECIFIC HEAT (J KG**-1 K**-1)	136.00	143.00	152.00	165.00	183.00	210.00
ALPHA,EXPANSION (E-6M M**-1 K**-1)	3.40	4.80	5.70	6.60	7.50	8.40
YMOD,TENSILE MODULUS (GPA)	410.00	350.00	365.00	340.00	310.00	270.00
YC , YIELD STRENGTH (MPA)	150.00	125.00	75.00	45.00	25.00	13.00

SPECIAL CASE FOR CANDIDATE IR AT 3 TEMPERATURES

T , TEMPERATURE (K)	273.00	773.00	1273.00
TCON,T. CONDUCTIVITY (W M**-1 K**-1)	147.00	110.00	74.00
CP , SPECIFIC HEAT (J KG**-1 K**-1)	130.00	142.00	159.00
ALPHA,EXPANSION (E-6M M**-1 K**-1)	6.80	7.20	7.80
YMOD,TENSILE MODULUS (GPA)	517.00	465.00	414.00
YC , YIELD STRENGTH (MPA)	234.00	234.00	43.00

SPECIAL CASE FOR CANDIDATE FE AT 6 TEMPERATURES

T , TEMPERATURE (K)	273.00	523.00	773.00	1023.00	1273.00	1523.00
TCON,T. CONDUCTIVITY (W M**-1 K**-1)	84.00	60.00	45.00	30.00	30.00	25.00
CP , SPECIFIC HEAT (J KG**-1 K**-1)	430.00	535.00	645.00	1075.00	715.00	715.00
ALPHA,EXPANSION (E-6M M**-1 K**-1)	12.00	14.00	16.00	17.00	23.00	23.00
YMOD,TENSILE MODULUS (GPA)	208.00	200.00	190.00	180.00	170.00	160.00
YC , YIELD STRENGTH (MPA)	280.00	175.00	130.00	50.00	20.00	10.00

- B. The bulk temperature rise in the coating layer due to the deposition of energy from a single pulse. The bulk temperature rise for this design should be minimized so as to reduce the stresses at the bond between the structural wall and the coating.
- C. The material frequency associated with the first wall due to the pulsed energy load. Any mismatch in frequencies between the coating and the structural wall will result in the weakening of the bond due to the induced tensile forces set up by the vibrational motions.
- D. The maximum first-cycle strain in the coating layer due to the motion of the cavity wall. This parameter represents the mechanical strains induced by the motion of the wall independent of any thermal effects; it should be minimized.
- E. The thermal stress in the coating layer due to the temperature gradient. The thermal stress levels relative to the yield strength of the coating indicates the integrity of the material to remain attached to the wall.
- F. The evaporation rate of coating material at/near the melting temperature. This parameter indicates the ease with which it is possible to remove pellet material which has accumulated on the wall as the device is pulsed.

The six parameters indicated above are calculated using the following expressions:

- A. Maximum temperature rise, ΔT_m ,

$$\Delta T_m = \frac{H_W}{S_A} \left(\frac{\pi}{4} \tau_p k \rho C_p \right)^{-1/2}$$

where H_W is the total first wall heat load, S_A is the first wall area, k is the thermal conductivity, ρ is the mass density, C_p is the specific heat, and τ_p is the pulse length of the heat load.

B. Bulk temperature rise, ΔT_B ,

$$\Delta T_B = H_W / (S_A h \rho C_p)$$

where h is the thickness of the coating layer.

C. Natural frequency of material x , ω_x ,

$$\omega_x^2 = 2 E_x / (\rho_x R_0^2 (1 - \nu_x))$$

where E_x is the tensile modulus, R_0 is the cavity radius, and ν_x is Poisson's ratio.

D. Maximum first-cycle strain, ϵ_m ,

$$\epsilon_m = ((1 - \nu_s) / (2 \rho_s E_s)) \times (I_r / h_s)$$

where the subscript s refers to the first wall structural material, and I_r is the reflected pressure impulse to the microexplosion, which, in SIU, is given by ⁽³⁾

$$I_r (\text{MPa s}) = 150 (E_{DT} (\text{MJ}))^{0.8} (2 R_0 (\text{m}))^{-1.39}$$

where E_{DT} is the total fusion energy per pulse in MJ and R_0 is the cavity radius in m.

E. The thermal stress in material x , S_x ,

$$S_x = 0.5 \alpha_x E_x \Delta T / (2(1 - \nu_x))$$

where α is the linear expansion coefficient, and ΔT is the temperature drop expected across the coating layer.

F. The evaporation rate, $\dot{m}_E$, at temperature T ,

$$\dot{m}_E (\text{g/s}) = 0.0583 P_V (\text{torr}) S_A (\text{m}) (A(u)/T(\text{k}))^{1/2}$$

where P_V is the vapor pressure, A is the atomic mass of the coating material, and the values of P_V are calculated using the following expression

$$\log P_V = A - \frac{B}{T}$$

for which the constants A and B are given in Reference 2.

In the conversion of these parameters to dimensionless magnitudes ranging from 0 to 1, each parameter was divided by an appropriate upper limit. This ratio was either used directly or subtracted from unity so that the final value was close to unity if meritorious or close to zero if not meritorious. For example, the maximum temperature rise was divided by the difference between the melting temperature T_m and the operating temperature T_o , and then subtracted from one, i.e.,

$$\text{FOM}(1) = 1 - \frac{\Delta T_m}{T_m - T_o}$$

where $\text{FOM}(1)$ is set to zero if $\Delta T_m > (T_m - T_o)$. Thus the further the coating material surface is from melting, the higher the value of $\text{FOM}(1)$. Similarly,

$$\text{FOM}(2) = 1 - \frac{\Delta T_B}{0.1 T_o}$$

where it is assumed that the bulk temperature rise should remain within 10% of the operating temperature;

$$FOM(3) = 2 \text{ MIN } [\omega_c, \omega_s] / (\omega_c + \omega_s)$$

where s refers to the structural material and c is the coating material, and $\text{MIN } [\omega_c, \omega_s]$ is the minimum of ω_c and ω_s ;

$$FOM(4) = 1 - \frac{Y_{MOD}}{1 - \nu_c} \frac{\epsilon_m}{Y_c}$$

where Y_c is the yield strength of the coating material and Y_{MOD} is the tensile modulus of the coating material;

$$FOM(5) = 1 - \frac{S_c}{Y_c}$$

and

$$FOM(6) = \dot{m}_E / (100 m_p r_p)$$

where m_p is the mass of a pellet, r_p is the injection rate (number/second), and it is assumed that a pulse causing melt and evaporation is injected every 100 pulses (on the average).

An overall figure-of-merit, $FOM(7)$, is assumed to be a weighted average of the above six FOM, where

$$FOM(7) = \sum_{i=1}^6 W_i FOM(i)$$

and

$$\sum_{i=1}^6 W_i = 1$$

For this first application of CANDID, four sets of W_i values were used. One set has all values equal to 0.1667. The second set gave more weight to the mechanical responses of the materials so that $W_4 = W_5 = 0.3$ but $W_1 = W_2 = W_3 = W_6 = 0.1$. In a third set, the melting characteristics were emphasized where $W_1 = W_6 = 0.3$ and $W_2 = W_3 = W_4 = W_5 = 0.1$. In the fourth set, the bulk temperature rise and relative frequency of wall materials due to reflected pressure impulses were ignored ($W_2 = W_3 = 0$), and $W_1 = W_4 = W_5 = W_6 = 0.25$.

4.3 Resulting Figures-of-Merit Values

The CANDID code was used to obtain the component FOM values for the 20 candidate materials. The results are shown in Table 4-6 for these materials which are all pure metals. Zeros indicate a failure of the material, e.g., aluminum melted since $FOM(1) = 0.0$. Failure in $FOM(2)$ indicated the bulk material temperature rises more than 10% of the operating temperature value of 773K assumed for the first wall. If $FOM(4) = 0$, the "cold" stress due to mechanical motion exceeds the yield strength, and if $FOM(5) = 0$ the thermal stress due to a temperature gradient of 50K exceeds the yield strength. The assumptions associated with the operation of the ICF reactor are summarized in Table 4-7. To evaluate $FOM(6)$ an arbitrary value of 1 gram was chosen for the mass of each pellet. This value was chosen to permit $FOM(6)$ to vary over the full range from near zero to unity for the 20 materials.

The tabulation of composite FOM values and ranking for each of the four weighting schemes as well as the average for all four is shown in Table 4-8. With only a few exceptions the value (V) and ranks (R) for the last pair of columns (set 5) are representative of all the various schemes. Aluminum, silver, copper, and vanadium ranked uniformly low, while rhenium, molybdenum, and chromium ranked uniformly high. Iridium and tungsten ranked high in all but the FOM which emphasizes the mechanical characteristics. The candidate selected in previous studies, tantalum, was found to be ranked at a consistent seventh place (see Table 4-8).

TABLE 4-6
COMPONENT FIGURES-OF-MERIT VALUES (V) AND RANKINGS (R)

Material	FOM(1)		FOM(2)		FOM(3)		FOM(4)		FOM(5)		FOM(6)	
	V	R	V	R	V	R	V	R	V	R	V	R
Al, Aluminum	0	11	.32	15	.99	2	.15	18	0	17	0	20
Ag, Silver	0	11	.33	14	.75	17	.57	13	0	17	.07	11
Au, Gold	0	11	.33	13	.60	20	.89	2	.81	6	0	17
Be, Beryllium	.12	9	.53	2	.61	19	.72	10	.44	15	.26	7
Cr, Chromium	.13	8	.50	5	.93	8	.54	14	.66	13	1.00	1
Cu, Copper	.18	6	.52	3	.92	11	0	19	0	17	.01	15
Fe, Iron	0	11	.51	4	.98	6	.80	5	.71	9	.54	5
Ir, Iridium	.56	3	.44	9	1.00	1	.38	15	.49	14	1.00	1
Mo, Molybdenum	.58	2	.42	11	.90	12	.75	7	.95	1	.19	8
Nb, Niobium	.17	7	.29	16	.88	13	.83	4	.85	4	.02	14
Ni, Nickel	0	11	.61	1	.98	5	0	19	0	17	.09	10
Pt, Platinum	.01	10	.42	10	.76	16	.72	9	.70	11	.01	16
Re, Rhenium	.55	4	.46	6	.92	9	.80	6	.84	5	.90	3
Si, Silicon	0	11	0	19	.80	15	.73	8	.91	3	.02	13
Ta, Tantalum	.36	5	.29	17	.85	14	.69	12	.74	8	.16	9
Th, Thorium	0	11	0	19	.69	18	.86	3	.80	7	0	18
Ti, Titanium	0	11	.45	8	.92	10	.71	11	.71	10	.07	12
V, Vanadium	0	11	.46	7	.99	3	.24	16	.12	16	.32	6
W, Tungsten	.70	1	.37	12	.99	4	.20	17	.67	12	.60	4
Zr, Zirconium	0	11	.13	18	.93	7	.91	1	.94	2	0	19

TABLE 4-7
INPUT DATA TO CANDID ON THE ICF DEVICE
AND STRUCTURE FIRST WALL

RO, ICF REACTOR CHAMBER RADIUS (m)	10.00
HO, CHAMBER WALL THICKNESS (m)	.0200
H, 1ST WALL COATING THICKNESS (m)	.0010
TO, 1ST WALL OPERATING TEMP. (K)	773.00
EOT, ENERGY PER PELLET EXPLOSION (MJ)	350.00
RATE, DROP RATE OF PELLETS (Hz)	10.00
PMASS, PELLET MASS (g)	1.0000
WLOAD, HEAT LOAD TO COATING (MJ)	160.00
GTEM, CAVITY GAS TEMPERATURE (K)	832.00
FATM, FRACT ABOVE TM FOR MELTING	.0200
PLEN, ENERGY PULSE LENGTH (s)	6.500E-05
GPRES, CAVITY GAS PRESSURE (torr)	1.000E-04
RHOC, DENSITY OF WALL MATL (gm/cm ⁻³)	7.70
POISC, POIS RATIO FOR WALL MATL	.2650
YMOCC, YOUNGS MODULUS OF WALL (GPa)	175.00
YOC, YIELD STRENGTH OF WALL (MPa)	340.00
TDROP, TEMP DROP ACROSS COATING (K)	50.00
TDMAX, TEMP DROP MAX RELATIVE TO TC	.1000
TCONC, T. COND. OF WALL (W M ⁻¹ K ⁻¹)	29.00
CPC, SP. HEAT OF WALL (J kg ⁻¹ K ⁻¹)	725.00
ALPHAC, WALL EXPANSION (10 ⁻⁶ K ⁻¹)	11.30
STO, WALL TENSILE ST. (MPa)	480.00
RPIO, REF. PRES. IMP. CONST.	150.00

TABLE 4-8
OVERALL FIGURES-OF-MERIT VALUES (V) AND RANK (R)
FOR FIVE WEIGHTING SCHEMES

Material	Set 1		Set 2		Set 3		Set 4		Set 5	
	V	R	V	R	V	R	V	R	V	R
Al, Aluminum	.24	20	.18	18	.15	20	.04	19	.16	20
Ag, Silver	.29	17	.28	17	.19	19	.16	17	.24	17
Au, Gold	.44	12	.60	7	.26	14	.42	10	.43	11
Be, Beryllium	.45	11	.50	15	.35	8	.39	13	.42	12
Cr, Chromium	.62	4	.61	6	.60	4	.58	4	.61	4
Cu, Copper	.27	19	.16	20	.20	17	.05	18	.18	18
Fe, Iron	.59	5	.66	4	.46	6	.51	6	.56	6
Ir, Iridium	.64	2	.56	12	.70	2	.61	3	.63	2
Mo, Molybdenum	.63	3	.72	2	.53	5	.62	2	.63	3
Nb, Niobium	.51	8	.64	5	.34	9	.47	8	.49	8
Ni, Nickel	.28	18	.17	19	.19	18	.23	20	.18	19
Pt, Platinum	.44	13	.55	13	.26	13	.36	15	.41	14
Re, Rhenium	.74	1	.77	1	.74	1	.77	1	.76	1
Si, Silicon	.41	14	.57	9	.25	15	.41	12	.41	13
Ta, Tantalum	.51	7	.59	8	.41	7	.48	7	.50	7
Th, Thorium	.39	15	.57	11	.24	16	.42	11	.40	15
Ti, Titanium	.48	10	.57	10	.30	10	.37	14	.43	10
V, Vanadium	.36	16	.29	16	.28	12	.17	16	.28	16
W, Tungsten	.59	6	.53	14	.61	3	.54	5	.57	5
Zr, Zirconium	.48	9	.66	3	.29	11	.46	9	.48	9

Of the 20 candidate materials investigated, 11 were found to have zero FOM values in one or more of the component FOM. Aluminum, silver, gold, copper, nickel, silicon, thorium, and zirconium have two or more zeros and were dropped as potential first wall coating candidates. The three materials, iron, titanium, and vanadium, have one zero value each and were also dropped. Note that although iron has a zero FOM component, the remaining values permitted the material to achieve high scores overall. Because beryllium is such a low-Z material it was also excluded. Even though platinum has no zeros, it has an overall ranking which is low and has a near zero FOM(1) value, and it was not included in the final selection.

The materials which remain as candidates are presented below in order of FOM ranking along with their associated atomic charge, and melting points

- A. Rhenium (Re), $Z = 186$, $T_m = 3453$ K
- B. Iridium (Ir), $Z = 193$, $T_m = 2720$ K
- C. Molybdenum (Mo), $Z = 96$, $T_m = 2883$ K
- D. Chromium (Cr), $Z = 52$, $T_m = 2148$ K
- E. Tungsten (W), $Z = 184$, $T_m = 3683$ K
- F. Tantalum (Ta), $Z = 181$, $T_m = 3269$ K
- G. Niobium (Nb), $Z = 93$, $T_m = 2741$ K

Based on the requirement of a high-Z material and a high melting point, chromium would likely be the first to be dropped when further studies are made, but since chromium is one of the more common materials relative to the list of seven candidates, it may have many merits worth considering. Four of the candidates (Re, Ir, W, Ta) have very high Z- values as well as high T_m -values, but considerations of material availability and manufacturability may make all four not viable candidates. The remaining materials, molybdenum and niobium, are only moderately-high-Z materials making them of limited value in terms of pellet materials. For only five of the seven metals listed above was there sufficient data on the temperature dependence of physical and mechanical properties to permit the calculation of FOM values at elevated temperatures. Values at room temperature, 500°C, and 1000°C are shown in

Table 4-9. In all cases the values decrease with increasing temperature with a character differing for each metal. Above 1000°C all five materials have zero values for at least one component FOM.

In addition to the FOM calculated for the materials, a number of important characteristics for each of the final seven candidates were collected and organized based on material given in Reference 3. The following is a summary of that information for each candidate:

RHENIUM (Re)

Main uses - electrical contacts, thermocouples, filaments for electronic devices, and alloying with Mo and W to increase ductility.

Special properties - generation of a nonpoisonous vaporous oxide occurs when heated in air at about 600°C.

Chemical properties - good resistance to corrosion.

IRIDIUM (Ir)

Main uses - crucible material, alloying with Pt for improved hardening, and electrodes and thermocouples.

Chemical properties - the most corrosion resistant element.

Fabricability - can be arc melted and e-beam melted and hot worked with procedures similar to those used for W; has limited malleability at room temperature.

MOLYBDENUM (Mo)

Main uses - electrical and electronic parts, high-temperature applications, corrosion-resistant equipment.

TABLE 4-9
OVERALL FIGURES-OF-MERIT VALUES VS. TEMPERATURE

Candidate	~20°C	500°C	1000°C
	~300K	773K	1273K
Mo, Molybdenum	.63	.52	.49
Nb, Niobium	.51	.50	.28
Ta, Tantalum	.51	.32	.31
W, Tungsten	.59	.54	.45
Ir, Iridium	.64	.66	.49

Special properties - not suitable for continued service at $\geq 500^{\circ}\text{C}$ in an oxidizing atmosphere.

Chemical properties - relatively inert in hydrogen, ammonia, and nitrogen up to $\sim 1100^{\circ}\text{C}$.

Fabricability - conventional methods are suitable for forming, but must be heated to proper temperature relative to its thickness and forming speed; cannot be hardened by heat treatment but only by work hardening; can be brazed or joined mechanically; can be welded by arc, resistance, percussion, flash, or e-beam methods.

CHROMIUM (Cr)

Main uses - alloying agent.

Chemical properties - good corrosion resistance.

TUNGSTEN (W)

Main uses - alloying for applications requiring high-density and/or high melting-point material; filaments.

Chemical properties - good corrosion resistance.

Manufacturability - very brittle as a pure metal; machinable, fairly ductile tungsten-based alloys have been developed, e.g., tungsten-nickel-copper alloys have good mechanical properties, fair ductility, and good machinability.

TANTALUM (Ta)

Main uses - chemical process equipment; heating elements.

Special properties - ductility is reduced by increased amounts of O_2 , N_2 , C, and H_2 .

Chemical properties - oxidizes in air at 300°C, excellent resistance to corrosion, embrittled by hydrogen.

Manufacturability - excellent using conventional methods, equipment and tooling; good joining characteristics.

NIOBIUM (Nb)

Main uses - superconductor; alloying element.

Special properties - oxidizes rapidly above 400°C; contamination by interstitial elements results in loss of ductility.

Chemical properties - moderate to high resistance to corrosion.

Manufacturability - conventional sheet-metal and tube working processes can be applied; weldable.

As indicated for a number of the above pure metal candidates, problems in manufacturability or loss of ductility could eliminate their applicability. As a potential means of reducing such problems so that such candidates remain attractive, alloys with the candidate pure metal as a base should be considered.

- A. Mo-Re and W-Re
- B. Pt-Ir
- C. Mo-W, Mo-Ti, and TZM
- D. W-Ni-Cu and W-Ni-Fe
- E. Ta-W and Ta-Hf
- F. Nb-Zr and Nb-W-Zr

Both rhenium and iridium are scarce and thus very costly, and the most likely use for either metal may lie in the addition of small amounts to materials such as molybdenum, tungsten, chromium, and platinum. The benefits of adding small amounts of rhenium are as follows:

- A. Solid solution strengthening
- B. Reduction in hardness
- C. Improvement in ductility
- D. Resistance to H₂ embrittlement
- E. Resistance to oxidation and corrosion

4.4 Summary and Recommendations

Using the CANDID code, 20 pure metals were investigated for potential application as first wall coatings for ICF reactor chambers. The data base of material properties included 19 physical parameters for each of the 20 candidate metals. Based on a number of these parameters as well as on several key ICF reactor parameters, six figures-of-merit (FOM) were calculated using CANDID for each candidate. By taking weighted sums of the six FOM values, overall FOM values were obtained. The six FOM calculations are based on mechanical, thermal, and vacuum vaporization engineering constraints. The 20 pure metal candidates investigated were aluminum, silver, gold, beryllium, chromium, copper, iron, iridium, molybdenum, niobium, nickel, platinum, rhenium, silicon, tantalum, thorium, titanium, vanadium, tungsten, and zirconium.

The candidate materials recommended for further consideration as first wall coatings are

- A. Rhenium (Re)
- B. Iridium (Ir)
- C. Molybdenum (Mo)
- D. Chromium (Cr)
- E. Tungsten (W)
- F. Tantalum (Ta)
- G. Niobium (Nb)

where the list is in rank order based on FOM values. Tantalum is the material previously selected for the ICF dry wall design. Based on the relative rankings of these seven materials in each of the six areas where FOM values were calculated, the following observations are made:

- A. W and Mo are especially suited for resisting surface melting (FOM[1]).
- B. Cr and Re are excellent candidates for maintaining small temperature changes at the interface with the structure wall (FOM [2]).
- C. Ir and W are the most compatible of the seven in terms of matching the natural frequency of the structural wall (FOM [3]).
- D. Re, Mo, and Nb are the best candidates in terms of withstanding both mechanical and thermal loads (FOM(4) and FOM(5)).
- E. Ir and Cr are particularly suited for wall conditioning by evaporation at elevated temperatures (FOM(6)).

Some basic weaknesses in the properties of the seven candidates include:

- A. Low FOM in performance under pulsed loading for Ir, Cr, and W.
- B. Low FOM in the matching of natural frequencies with that of the structural wall for Ta and Nb.
- C. Low FOM in bulk temperature rises for Ta and Nb.
- D. Low evaporation rates at elevated temperatures for Nb.

Overall, rhenium has component FOM values which are relatively high in all areas investigated. Molybdenum has FOM values which are good to very good in all areas and has no areas with low FOM values. All the other candidates show weaknesses in one or more of the areas examined. The candidate selected for the ICF dry wall study i.e., tantalum, continues to show promise, but there

are areas of potential weakness in that three of the six FOM values placed Ta in the lower half of the ranking when considering all 20 candidates. Since the six FOM chosen to characterize the candidate metals are limited to thermal, mechanical, and vacuum considerations, it is recommended that additional FOM be selected and used in future work. These could include cost, fabrication, irradiation effects, compatibility with pellet design, etc.

In collecting temperature-dependent data for this study and for CANDID input, very little high temperature data was found. For five candidates (Mo, Nb, Ta, W, and Ir) enough data was collected to permit a limited temperature-dependent study. The FOM values were found to be strong functions of temperature, and were found to have changes with temperature which do not follow any characteristic pattern for the five materials studied. At the highest temperature (1273 K) all FOM values were much lower than at 20°C, indicating the importance of using data at the elevated temperatures expected in the ICF reactor.

In accumulating the material properties data a number of publications were used. In a large number of cases, published results were consistent when comparing properties listed in different publications, but there were many cases where substantial differences occurred. For example, in collecting values for tensile strengths and yield strengths (see Tables 4-1 to 4-5) from References 1 and 2, magnitudes differed by as much as a factor of two. In further work, one should take a more careful look to be sure such inconsistencies are cleared up. In addition, a number of the values for material properties apply to test samples which have undergone processes which were different than will be undergone for the first wall coating material. Since properties are generally process-dependent, further considerations will require more careful analyses with respect to such matters.

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5.0 RESPONSE OF TANTALUM COATING TO PELLET YIELD IRRADIATION

The novel dry wall concept for protecting the structural wall of an ICF reactor utilizes a thin layer of high Z, high melting point metal, Ta, to buffer and protect the steel first wall. This protection concept offers the advantages of simplicity in construction and operation and of compatibility with both laser and heavy ion drivers. The first wall is composed of thin steel tubes carrying flowing liquid lithium to remove the steady state surface heat and is protected by a layer of Ta whose melting temperature is above the peak temperature transient caused by the x-rays or ions. This coating buffers the steel tube from the large temperature transients which might otherwise destroy it due to thermal stress cyclic fatigue. This coated thin tube wall design handles time averaged fusion thermal power of 3500 MWt in a chamber at a distance 10 m from the explosion. The neutron energy is absorbed in the first wall coolant and in the thick flowing liquid lithium blanket situated behind the tubular first wall. Tritium breeding ratio for this configuration is about 1.22.

The viability of this dry wall concept was confirmed during the performance of the previous contract⁽¹⁾ by three sets of calculations using the CHART D radiation transport and materials code. The first calculation demonstrated that the maximum transient temperature increase at the Ta surface due to x-ray and/or ion absorption was some 400°C below the melting point of Ta. The second calculation demonstrated that a layer of about 1 mm of Ta is sufficient to reduce the temperature transient at the Ta-steel interface to very low values, of the order of 50°C. These two calculations confirm that a coherent Ta coating will indeed buffer the steel wall from high thermal transients without being destroyed by evaporation or ablation. However, the large temperature gradient across the coating may generate internal stresses large enough to destroy the coating by spallation or flaking. The third calculation explored this possibility and found that stresses connected with this would be relieved by plastic flow which would likely occur to a depth of several μm .

Examination of the interaction between pellet heavy metal debris ions and the first wall showed that, no matter what the original first wall material was, the rate of condensible metal deposition on the first wall was so great that the first wall coating material must be considered to be the same as the pellet metal. The choice of heavy metal for the pellet therefore becomes a joint decision of the pellet designer and the reactor designer. Candidate heavy metals were reviewed and Ta was selected because of its high melting temperature, ease of handling, ductility, and general acceptability as the target's heavy metal component.

However, the original CHART D calculations which had been used to confirm the viability of the dry wall concept had been based on data for a pellet whose heavy metal was not tantalum. Los Alamos redid the calculations for a pellet using Ta as the heavy metal.

The CHART D calculations identified temperature and stress conditions at the surface and within the Ta coating as well as failure of the surface by spallation. However, for conditions below failure, processes such as evaporation or sputtering may introduce significant losses from the coating. Section 5.3 uses computer codes developed at Los Alamos to calculate the magnitude of these effects.

Inside the ICF reaction chamber, Ta will be deposited at some rate onto the original coating. In order to prevent too large a buildup of Ta, some removal process, either continuous or intermittent, must be introduced. Section 5.4 explores the possibility of equilibrating Ta deposition rate with Ta loss rate by adjusting pellet yield, chamber radius and pellet output spectra.

In the previous work two simplifying assumptions were made: (1) the pellet debris ions (H, D, T, O, C, Si, Ta, etc.) which strike and penetrate the Ta do not change the physical or chemical properties of the Ta coating, and (2) the presence of background gas has no effect on the dynamics of evaporation or deposition, Section 5.1 explores these assumptions.

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5.1 Chemical Reactions Between Pellet Debris and Tantalum

The reaction between tantalum and the debris from the pellet explosion must be considered in relation to the depth of implantation of the debris ions and the rate of implantation. This is necessary because the energetic ions are physically implanted into the Ta, and the subsequent chemistry will depend upon the concentration of the implanted species and the temperature in the Ta wall. Because of the high wall temperature it is expected that the implanted ions will attain rapid chemical equilibrium with the Ta.

As a first approximation the pellet debris is considered to be present only as monatomic ions. If all of the ions impinge onto the wall, there will be approximately 10^{12} ions/mm²/shot. At a repetition rate of 10 Hz there will be approximately 10^{13} ions/mm²/second. The atomic density of the tantalum wall is approximately 5×10^{19} atoms/mm³. If the Ta is divided into sections of 1 μ m thickness, each section will contain approximately 5×10^{16} atoms/mm². A 1% concentration of debris in this section will be approximately 5×10^{14} atoms/mm²; this concentration will be attained in ten seconds of debris implantation at 100 percent implantation.

At an ion energy of 5 keV/amu the depth of penetration of hydrogen will be extremely shallow ($<10^{-3}$ μ m).⁽¹⁾ The heavy ions, such as those of Ta and Si, may penetrate to depths of ~ 1 μ m. It is in this context that the reactions of the tantalum wall with each debris species must be considered.

5.1.1 Ta - H, D, T Reactions

Tantalum forms the hydride, TaH₂, at low temperature, but this compound is unstable at the drywall temperatures. However, hydrogen has a significant solubility in Ta, with the solubility depending upon the temperature and the pressure of H₂ over the solid, as shown in Figure 5-1.⁽²⁾ As seen from this figure, the H₂ solubility will be extremely low at temperatures above 600 C and pressures of <0.1 torr. When the tantalum also contains Si, C, and O, the solubility of hydrogen will be further decreased because some of the hydrogen sites will be occupied by the other impurities. The solubility of H in pure Ta may be calculated by the equation:⁽³⁾

Fig. 5-1. Solubility of H in Ta (Ref. 2).

$$\ln P_{(\text{Torr})}^{1/2} = 9.970 + 1.0031 \ln\left(\frac{r}{0.71 - r}\right) + \frac{1}{T} (-4051.4 - 3334r + 8351r^2 - 17057r^3 + 16.951r^4) \quad (1)$$

where $r = \text{H/Ta atom ratio}$, P is the equilibrium H_2 pressure over the solid, and T is the absolute temperature. At temperatures above 600C and $P < 0.1$ torr, r is < 0.01 , so that the higher order term of r may be neglected. At 1100 K, and $P \sim 10^{-4}$ torr, $r \sim 10^{-5}$. As noted above, the hydrogen ion penetration into the tantalum will be very shallow and the average temperature of the wall to a depth of approximately 25 μm will be at least 1100 K.⁽⁴⁾

Because the penetration of the debris ions into the tantalum wall is expected to cause high concentration gradients at depths up to 1 μm , it is of interest to consider the subsequent diffusion of each implanted species into or out of the wall. Table 5-1 summarizes the diffusion coefficients of each element in pure tantalum. It is not known exactly how the presence of the different impurities will affect the diffusivities of each species in the tantalum.

As seen in Table 5-1, the diffusivity of H in Ta is extremely fast. In view of the high wall temperature and high diffusivity of H, and the low chamber pressure and shallow hydrogen ion penetration, it is reasonable to state that the accumulation of hydrogen in the tantalum wall will be very low over the long term. The low concentration of dissolved hydrogen should have no adverse effect on the mechanical properties of the tantalum.

5.1.2 Ta - C Reactions

Tantalum forms two stable carbides of formulae Ta_2C and TaC . The solid solubility of carbon in Ta is 0.25 atom percent up to 1500C. Figure 5-2 shows the phase diagram for the Ta-C system.⁽⁸⁾ As seen from the diffusivity for carbon in Table 5-1, the diffusion of carbon into the tantalum wall after ion implantation will be a relatively slow process. The carbon, which could accumulate at the rate of approximately 10^{13} atoms/ $\text{mm}^2/\text{second}$, therefore will form the stable carbides on the tantalum surface. The

TABLE 5-1
DIFFUSION COEFFICIENTS FOR VARIOUS ELEMENTS

$$D = A \exp\left(\frac{-Q}{RT}\right), \text{ in cm}^2/\text{sec at } T > 800 \text{ K}$$

<u>Diffusing Element</u>	<u>Diffusivity, A, cm²/sec</u>	<u>Q, cal/mole</u>		<u>Ref.</u>
H	1560	32,230	at 5 atom % H	5
H	13960	33,620	at 10 atom % H	5
O	0.01	27,000		6
C	0.0067	38,600		7
Si	0.036	21,150		7

Fig. 5-2. Phase Diagram for the Ta-C System (Ref. 8).

thermodynamic stability of the carbides precludes the vaporization of carbon via reaction with oxygen or hydrogen. If the penetration of the carbon ions is approximately $1\text{ }\mu\text{m}$, this surface will become rapidly saturated with the carbon. However, because there is also a simultaneous implantation and surface deposition of Ta debris, the implanted carbon will be completely reacted to form tantalum carbide.

5.1.3 Ta - Si Reactions

As with carbon, silicon also forms several stable silicides, as shown in Figure 5-3.⁽⁹⁾ The solid solubility of Si in Ta also is very low, so that the silicides will be formed upon silicon ion implantation. As with C, the Si also will accumulate near the surface of the wall because of the low diffusivity, and because the silicon will not form a volatile compound with hydrogen or oxygen.

5.1.4 Ta - Si, C Reactions

It is important here to consider the nucleation and formation of discrete crystallites of the carbides and silicides as a result of ion implantation and subsequent reaction. Ion implantation at low temperatures results in the production of nonequilibrium states of the implanted system. However, the high temperatures attained near the surface of the tantalum wall will cause the reaction and equilibration of the implanted species. At temperatures above 1000°C there will be a high probability of crystallite nucleation of the carbides and silicides. If these crystallites should grow to large dimensions, there will be a high probability for crack formation in the tantalum wall as a result of the crystal anisotropy and of the temperature fluctuations, the latter due to the 10 Hz repetition rate. If the crystallites remain finely dispersed as microcrystals, the tantalum wall will become appreciably hardened. Clearly, experimental data are needed to determine the nucleation and growth rates of the crystallites, and the effect of these crystallites on the mechanical integrity of the tantalum wall.

Fig. 5-3. Phase diagram for the Ta-Si System (Ref. 9).

Fig. 5-4. Oxygen Solubility in Tantalum as a Function of Temperature (Ref. 10).

5.1.5 Ta - O Reactions

Extensive studies have been made of the Ta-O system, and Figure 5-4 shows the oxygen solubility.⁽¹⁰⁾ The more recent data of Kofstad⁽⁶⁾ appear to be the most accurate. Oxygen has a solid solubility in Ta which is appreciably higher than those of carbon and silicon in the same metal. However, the diffusivity of oxygen in Ta also is relatively low. When the solubility limit of oxygen is exceeded at any given temperature the oxygen then forms the stable oxide, Ta₂O₅. If this latter oxide accumulates on the surface and forms a film it will tend to spill off; whereas the carbide, and silicides tend to form very adherent films on the tantalum.

If the temperature of the oxygen - impregnated surface is higher than 1600 C, the surface material will start to vaporize as TaO and TaO₂.⁽¹¹⁾ Turkdogan⁽¹²⁾ gives the evaporation rate, R, as $R - R_0 = k_1 C + k_2 C^2$, where R_0 is the evaporation rate of pure Ta in vacuum, C is the oxygen solubility in the metal, and $k_1 = 1.15 \times 10^5 \exp(-118,230/RT)$ and $k_2 = 1.60 \times 10^8 \exp(-132,590/RT)$ are rate constants for the formation and vaporization of TaO and TaO₂. At 1900 K (1627 C), the evaporation rate will be 2.3×10^{12} molecules/mm²/second. It is not known to what degree this evaporation rate will be affected by the presence of the carbides and silicides. However, if the wall temperature remains > 1600 C there will be loss of tantalum as the volatile oxide; in contrast, tantalum metal will not vaporize until the temperature is appreciably higher.

5.1.6 Ta Implantation

As discussed above, the carbides, silicides, and oxides are the only compounds formed as a consequence of the surface ion implantation. However, because the pellet is composed of a tantalum shell, Ta ions will also be embedded into the wall. The pellet will yield 21.4% of Ta,⁽⁴⁾ excluding hydrogen, and it may be assumed that nearly all of this metal will be condensed onto the surface by ion implantation. As shown above, the accumulation of debris elements on the wall can be very rapid. A large part of the implanted tantalum will also react with the implanted carbon, silicon, and oxygen to form the respective

compounds. The implantation of the debris ions will tend to form distortions in the tantalum substrate, and there also may be severe strains created in the surface film as a result of the high concentration of implanted material. Because of the complexity of the implanted surface, it is not possible to assess this strain which could result in surface fracture and spallation. Should the integrity of the surface be maintained, the wall will increase in thickness with continued implantation.

5.1.7 Gas Phase Reactions

Because of the extremely high energies at which the pellet debris will be formed, it may be assumed that, in addition to H^+ , D^+ , and T^+ , there will be multiply charged heavy positive ions and electrons in the plasma. The recombination of these ions before reaching the first wall should be negligible at chamber pressures of 10^{-3} to 10^{-4} torr.⁽¹³⁾ At high pressures, however, there will be some recombination of electrons and ions in the presence of a third body, and some multiply-charged ion-electron recombination may take place within the blast wave.

A reaction which must be considered is that between $TaO(g)$ and the debris ions. As discussed above, there will be evaporation of TaO from the hot surface of the wall. The kinetic energy of the TaO molecules, $3/2 kT$, will be negligible as compared to the ions of 5 keV/amu. The reaction, therefore, involves the collision of a highly energetic ion with a relatively stationary molecule. Under this condition, the collision will result in the dissociation of the TaO , via reactions such as,

Because of the number of different species involved, the number of possible reactions is large and complicated. If the Ta^+ should eventually arrive at the wall, the metal should condense by recombination with an electron on the surface. The secondary Ta^+ ion formed by collision with H^+ will have kinetic energy less than 5 keV, so that there should be almost no ion penetration. It should be noted that the mean free path of molecules in the chamber will be approximately one meter. The Ta in reaction (4) also may eventually condense on the wall after several collisions. The gas phase reactions, therefore, will tend to recondense the tantalum onto the first wall surface.

5.1.8 Summary of Ta Wall Reactions

- A. The accumulation of H, D, and T, and the formation of hydrogen bubbles on the first wall will be negligible.
- B. The ion implantation of the heavier debris ions will be approximately $0.5 \mu m$ at most.
- C. The carbon and silicon will form stable, non-volatile carbides and silicides via reactions with the Ta substrate and with implanted Ta.
- D. It is likely that the implantation of the debris will cause high surface stresses on the wall, with subsequent surface damage and spallation. But this must be investigated experimentally to determine the magnitude of the effect.
- E. In the absence of excessive spallation the first wall should increase, or maintain, its thickness via the accumulation of the debris.

5.1.9 Other Refractory Metal First Walls

As with tantalum, the refractory metals, W, Mo, and Nb also form thermodynamically stable carbides and silicides at high temperatures.⁽⁷⁾ Also, the solubility of hydrogen in these metals is very low at high

temperatures. However, the oxides of the latter metals have appreciably lower melting points and higher vapor pressures than those of tantalum.⁽¹⁴⁾

Because the debris will contain 13 to 15% O, the first wall deterioration may be more rapid with W, Mo, and Nb than with Ta.

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5.2 Physical Effects in the First Wall Coating

The design of ICF pellets for reactor considerations must provide a high-yield thermonuclear burn of the deuterium-tritium fuel contained within the pellet. The burn products--X-rays, charged particles, and neutrons--form intense pulses of surface and volumetric heat which must be removed from the first wall and the blanket/shield, respectively. For the dry wall design, the X-ray pulse heats the wall to near-melting temperatures while the charged particles making up the pellet debris do not heat the wall to as high temperatures. The soft X-rays are particularly difficult to handle because the associated pulse time is short making the pulses very intense. In addition, deposition profiles into the first wall are strongly peaked at the surface.

For the reference dry wall design the first wall coating is a 1-mm layer of tantalum metal on the HT-9 steel tubes making up the first wall. The pellet design also uses tantalum so that continued pulsing will result in the build up of the tantalum coating. Of course, all condensible debris products will contribute to the wall build-up and the fraction which is tantalum will depend on the pellet design. As a method to maintain the tantalum coating at or near a thickness of 1 mm, the concept of a special pellet⁽¹⁾ was developed. This pellet would burn such that the soft X-ray pulse would have sufficient power and pulse duration to reduce the built up material by a combination of evaporation and spallation. In this section of the study report, the results of code calculations on the interactions of X-ray pulses with the tantalum first wall coating are presented. The results are based on deposition profiles generated from pellet yields calculated at LANL. The profiles are required as input to the code calculations to specify the energy source term.

In the remaining subsections, the following aspects of the pellet/wall interactions are discussed: X-ray photoabsorption in tantalum, the CHART D code, and the calculational results.

5.2.1 X-ray Photoabsorption in Tantalum

In order to specify the energy source which is interacting and heating the 1-mm tantalum coating, the deposition of energy as a function of depth into the coating layer must be known. For X-radiation, photoabsorption in solids is the dominant mechanism for energy deposition for low-energy X-rays. In terms of cross sections, an extensive compilation of experimental data⁽²⁾ at energies of 1 keV and greater can be used. Figure 5-5 shows the data for tantalum taken from that compilation which show the effects due to the atomic structure of tantalum. In another compilation,⁽³⁾ a mathematical model was developed for calculating absorption coefficients, including the atomic shell effects, for X-ray energies between 1 and 50 keV. For energies below 1 keV, neither of these compilations are applicable.

For the low energy region, a new compilation⁽⁴⁾ has recently been published. This work includes tables previously compiled⁽⁵⁾ by the University of Hawaii as well as new tabulations covering the region from 30 eV to 300 eV. For reports on actual measurements of soft X-ray absorption in tantalum see Reference (6) and (7). For energies below 2 keV, the absorption coefficients are taken from Reference (4), and are shown in Figure 5-6. Any absorption in the very low energies (<30 eV) is assumed to be associated with the excitation of volume plasmon⁽⁸⁾.

The distribution of the source of X-rays is assumed to be that describing black body radiation. Tantalum, like most solids, absorbs the radiation entering through its surface very strongly with absorption occurring in a layer which is only a fraction of a micron in thickness. To obtain the intensity in each wavelength region a tabulation⁽⁹⁾ of monochromatic intensities for black body radiation in vacuum was used. The absorption coefficients integrated over this intensity distribution provides the deposition profile of the source X-rays. For the time history of the source, a four-point deposition was used where the first and last points are the time values for the start and stop of the pulse, respectively. The second and third points are the time values between which the source is at its full intensity. See Figure 5-7.

Figure 5-5. The Absorption Cross Section in Tantalum for X-rays at Energies Greater Than 1 keV (Ref. 2).

Figure 5-6. The Absorption Coefficient in Tantalum for Soft X-rays (Ref. 4).

Figure 5-7. The Temporal Behavior of the Reference X-ray Energy Pulse Used in CHART D.

5.2.2 The CHART D Code

The CHART D code was the main calculational tool used in the analysis of the pellet/wall interaction. The code provides a quantitative description in one-dimension of the hydrodynamic motion of a system when coupled to the transport of energy. The equation of state calculations in CHART D are considered to be outstanding. For the tantalum coating the parameters for the equation of state were supplied as external inputs to the code. Those parameters which were supplied are for the pure metal, even though the pellet debris will be composed of tantalum contaminated with other materials and having a composition similar to that of the pellet. It is clear from the results given in Section 5.1 that the parameters will be altered from those of the pure metal, but it is expected that for the Chart D analyses performed, the pure metal characteristics can be applied without jeopardizing the main conclusions of the study.

One of the key outputs of the CHART D calculations is the prediction and quantification of the spallation of the tantalum coating. The code output shows the formation of voids or gaps between layers of material, which were not originally present, at the time the tantalum is spalling. The spallation process is treated in two independent sets of calculations. The first is through the modeling of the fracture mechanics of the process. Three fracture models are actually employed as follows: 1) stress gradient model, 2) cumulative damage model, and 3) tensile strength model. In the first one, the temperature for the threshold of spallation is a function of the static tensile strength, the ultimate maximum strength, and the temperature at which the strength vanishes. In the cumulative damage model, material properties including static tensile strength are also used to establish the criteria for threshold. For the third model, the criterion is based on the maximum tensile strength and it is assumed that the static tensile strength is equal to the absolute maximum strength. The user sets the input to select one of these three models. In the calculations presented in this report the tensile strength model was used. The selection of this model was made because it provides the criterion most widely used and requires the least amount of materials data. Since the actual material coating is not pure tantalum metal,

this option seems the most logical to employ until more material and design information becomes available.

The strength used for tantalum is 180 MPa, and the strength vanishing temperature was set at the melting point (2996°C). The strength value is based on data for the elevated-temperature tensile properties of 1-mm thick electron-beam-melted tantalum sheet.⁽¹⁰⁾ This value assumes no stress relief or recrystallization.

The second independent set of calculations involving the spallation process is the more time consuming. These involve the motion and the related properties of the extra free surfaces in the interior of the material. Extra position and velocity values are calculated for each void. It is important to note that the formation of voids due to spallation occurs only at zone boundaries. These boundaries are defined by the space-time mesh employed by the code user in setting up the input to the code. For the present study, the zoning of the tantalum region was prepared with the soft X-ray deposition profiles in mind. In particular the front zones had thicknesses for which less than 10% of the total X-ray energy was deposited in any single zone. For that reason the first few zones had thickness on the order of a few thousandths of a micron. The zones were set up with the first one being the thinnest and increasing gradually so that the final zones behind the X-ray deposition were much thicker. Over the depth of tantalum which was active with X-ray absorption there were a total of 25 zones with thicknesses ranging from 0.002 μm to 0.02 μm .

5.2.3 Calculational Results

There were two main objectives to this phase of the study. In the first objective, CHART D was to be exercised in the reestablishment of a reference calculation for the pellet/wall interaction for the dry wall concept. For this reference case, the latest calculations of pellet yield data based on a pellet composition which included tantalum were used. The pellet parameters were adjusted until the corresponding soft X-ray deposition profile in the

first wall coating did not result in any evaporation or spalling. In the second objective, a deposition profile was to be used which caused the spallation in a controlled manner of several layers of tantalum. In this latter case, the pellet is the special one which will reduce the build up of Ta on the first wall by the spalling and subsequent removal from the pellet chamber of the spalled material.

The CHART D results for the temperature behavior of the coating as a function of time and depth are shown in Figures 5-8 and 5-9. The time characteristics of the X-ray source term are shown in Figure 5-7. As can be seen from the various curves in Figure 5-8, the response of the surface zones of the coating are immediate while the back zones have delayed responses. The temperature rise is the greatest in the initial phase of heating even before the pulse has reached its full energy level. The full energy level occurs at a pulse time of 15 ns, which is an estimated value of the time lapse between the initial surface heating and the peak surface heating. It is assumed that the ramp down time characteristics of the pulse also involve a 15 ns time period starting at a pulse time of 105 ns. It is at a pulse time of 105 ns that the surface begins cooling back down even though the pulse does not end until a time of 120 ns. The responses of the back zones of the coating are characteristically different in that the temperature rise is delayed beyond the pulse start times and does not end until after the 105 ns point in time. These characteristics are also shown in Figure 5-9. The differences in profile characteristics between times of 105 ns or less and times greater than 105 ns are particularly apparent in Figure 5-9.

The peak surface temperature was calculated by CHART D to be 0.2106 eV (2440 K). Using the analytical expression given in Section 4 for the maximum temperature rise, the value of the peak surface temperature was calculated to be 2410 K; this is excellent agreement with the CHART D value. The material parameters used in the hand calculation were for elevated temperatures. For room temperature parameters a value of 3000 K is calculated. The melting point of tantalum is at a temperature of 3269 K so that the peak temperature is only 75% of the melting point. Thus surface melting is unlikely. As a second check on the CHART D calculations, the analytical expression given in

Figure 5-8. The Temporal Behavior of the Zone Temperatures of Several Zones Characterized by the Depth, Δ , to the Back of Each Zone for the Reference Case Involving no Melting or Spallation of the First Wall (Melting Point of Tantalum is 0.2817 eV).

Figure 5-9. The Temperature Profiles as a Function of Depth into the First Wall Coating at Several Times, t , During the Reference Pulse Which Involves no Melting or Spallation. The Times, t , are Related to the Temporal Behavior Shown in Figure 3.

Section 4 for the bulk temperature rise, ΔT_B , was used to calculate a depth, h , into the coating. If ΔT_B is assumed to be half the peak temperature rise (i.e., half of 2440 K - 770 K), the expression yields a value of approximately 3 μm for h which is the point at which an extrapolation of the straight portion of the $t = 105$ ns curve in Figure 5-9 crosses the 0.066 eV (770 K) line. Thus, a combination of this result and the value for the peak surface temperature seems to provide a simple analytical means of estimating the temperature gradient near the surface and at the end of the pulse.

In addition to the temperature behavior of the tantalum during the X-ray pulse, the density behavior predicted by CHART D was carefully examined. The relative density (where a value of 1.0 corresponds to 16.6 g/cm³) as a function of pulse time and coating depth shown in Figures 5-10 and 5-11 for a few specific cases. The temporal behavior (Figure 5-10) shows a dramatic reduction in density in the first few nanoseconds followed by a more gradual density reduction until near the end of this pulse where the change is toward increasing densities. Again, as in the temperature dependence, the density begins to "recover" for the surface zones near $t = 105$ ns while the back zones begin the recovery later in time. As a function of depth for specific pulse times (see Figure 5-11) we see the reduced density moving into the coating with time and the reduction remaining at depths well beyond the first few microns. In fact, the relative density of 0.98 remains for approximately 40 μm and 260 μm beyond the surface for $t = 15$ ns and $t = 105$ ns, respectively, before returning to unity. These results are calculated based on the assumption that the back surface of the tantalum which is bonded to the HT-9 steel tubing is fixed and does not undergo any displacement during the pulse. This may be an over-simplification of the actual situation, but is a reasonable assumption at this point in the study since the HT-9 tubes are part of the structural system supporting the pellet chamber and should remain relatively motionless during time period of the soft X-ray pulse.

The front surface of the tantalum coating is not assumed to be fixed, so that the absorption of the X-rays will result in the motion of the front surface toward the center of the chamber resulting in a reduction in the coating

Figure 5-10. The Temporal Behavior of the Relative Density of the Tantalum Coating at the Surface ($\Delta=0$) and at a Depth of One Micron ($\Delta=1\mu$) When a Value of 1.0 Corresponds to 16.6 g/cm^3 . This is for the Reference Case.

Figure 5-11. The Relative Density of the Tantalum Coating as a Function of Zone Depth for a Time Corresponding to the Beginning of the Max Energy Deposition and a Time Corresponding to the End of the Max Energy Deposition for the Reference Case.

density. The front surface moves toward the chamber center at an average speed of approximately 47 m/s. The motion of the zones at a depth of 40 μm have similar velocities at $t = 15 \text{ ns}$; at $t = 105 \text{ ns}$, the depth extends to approximately 260 μm . These depths correspond to the points at which the relative density remains at a reduced value. The displacement of this front surface of the coating along with the depth of the penetration of the thermal energy into the coating are shown in Figure 5-12. The depth of the thermal penetration is a measure of the total thickness into the coating that the temperature has risen above the pre-pulse value of 0.0661 eV (770 K). As can be seen in examining the figure, both the displacement of the front surface and the depth of the thermal penetration have values which are close to each other through out the pulse. The motion of the first wall coating can thus be associated with the thermal expansion of the material as the energy pulse travels through. As the energy pulse ends, the penetration of this thermal energy slows, but the inertia of the first wall motion continues and even after 190 ns the speed is greater than 40 m/s.

In this subsection we discuss the CHART D calculations of a pellet/wall interaction which result in the spallation of the first few front zones of this tantalum coating. Several calculations were made in a search for cases in which the CHART D code predicted spallation followed by the "recovery" of the tantalum coating such that the spallation process ended and the wall began cooling down. Unfortunately, in all attempts, the calculations could not be carried out to that point. A number of computer operational errors were experienced and the calculations were abruptly terminated in all attempts. The calculations did progress to a pulse time very close to the end of the pulse and these results are discussed in the following paragraphs.

Shown in Figures 5-13 and 5-14 are the temperature characteristics as a function of time and coating depth, respectively, for a representative set of CHART D calculations for the case of the special pellet leading to the spalling of the first wall. These correspond to Figures 5-8 and 5-9 for the case of the reference pellet for which spallation did not occur. As can be seen in Figure 5-13, the temperature behavior is dramatically altered after a pulse time of 95 ns; this is the point at which the first zone spalled. As a

Figure 5-12. The Thermal Penetration Depth of Zones with Temperatures Above the Equilibrium Value (730K) and the Displacement Toward the Center of the Spherical Chamber of the Surface of the First Wall as a Function of Pulse Time.

Figure 5-13. The Temporal Behavior of the Zone Temperatures of Several Zones Characterized by the Depth, Δ , to the Back of Each Zone for the Case Involving Spallation and Vaporization. The CHART D Calculation Ended at a Pulse Time of 98.7375 ns.

Figure 5-14. The Temperature Profiles as a Function of Depth into the First Wall Coating at Several Times, t , During the Case in Which Spallation and Evaporation Occurred.

result of the spallation of zones, voids are formed causing thermal contact between the zones to be lost. As a consequence, the surface layers which are spalled retain the energy deposited by the X-rays and show a quick rise in temperature. On the other hand the zones behind the spallation become thermally isolated and actually show a dramatic fall in temperature. This is because the spalled layers are initially in a solid state and are not vaporized instantaneously. Vaporization of the first spalled zone is calculated in CHART D to occur 1 ns following spall. Even for the final time step printed for this computer run, which is for a pulse time of 98.74 ns, only five zones were predicted to have vaporized compared to a total of 19 zones spalled. The calculations indicated that there were also a number of times that a vaporized zone actually recondensed, for a brief period, back into the solid state and vaporized a second time. The change in temperature behavior as a function of coating depth due to the spalling of zones can also be seen in Figure 5-14 where the curve for $t = 98$ ns shows a very different behavior from that for $t = 90$ ns. The strong rise in temperature of the zones at the surface and the dip in temperatures just behind the surface zones is clearly displayed. This effect of the thermal redistribution of the surface region also causes the spalled zones to begin moving more rapidly toward the chamber center. In the reference case when spallation did not occur, the front zones had speeds near 43 m/s at $t = 98$ ns. For this case, the surface zones are moving at speeds of 85 m/s in a vaporized state at a temperature of 0.39 eV (4530 K) while the zones further back have speeds nearer to those of the reference case. Unfortunately there are no calculations beyond a 99 ns pulse time so that the behavior of the coating following the X-ray heating cannot be quantified.

One CHART D run was made in which the X-ray pulse was "shut off" at the instant of the first spallation and allowed to ramp down for 15 ns smoothly to zero energy flow. The code still terminated abruptly and calculated characteristics very similar to these presented in Figure 5-13 and 5-14.

As a final consideration, the dependence of the depth of the last zone spalled as a function of pulse time was examined. As can be seen from Figure 5-15, the spallation process is a very abrupt resulting in an almost instantaneous spallation of zones. The time difference predicted by CHART D between the two flat regions of the curve in Figure 5-15 is only 0.044 ns. It is not known if the second flat region marks the end of the spallation followed by a recovery of the coating to lower temperatures. If it does, a total of 2200 g of coating material was spalled. This result should not be considered as an accurate prediction of the amount of spalled material since the material properties of tantalum contaminated with pellet debris are not known, and since the CHART D calculation was not carried beyond the pulse. These results do indicate, however, a number of features of the spallation process and do indicate the potential for the use of a special pellet for the removal of pellet debris built-up on the first wall following a period of normal pulsing.

Figure 5-15. The Depth of the Spallation as a Function of Time for the Case in Which Spallation Occurred.

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6.0 MANUFACTURABILITY OF COMPOSITE WALL

6.1 Introduction

The Westinghouse ICF dry wall concept utilizes a thin (~1 mm) tantalum coating to protect the HT9 tubes comprising the reaction chamber first wall from the transient temperature increase due to pulses of low energy x-rays and ions. In order to provide protection, coating adherence must survive both the rapid cyclic thermal stresses due to the individual pellet microexplosions as well as the low cycle thermal stresses caused by plant startup and shutdown. In addition, the method of applying the coating should be compatible with ultimate commercial application. Some of the issues resulting from this latter requirement are:

- o Can the coating be applied in-situ rapidly and reproducibly
- o Can the Ta-steel system be manufactured in a reliable and cost-effective manner
- o What is the most promising method of manufacture

The following sections describe our approach to resolving these issues.

6.2 Coating Method Selection

Three coating techniques were examined for the present application:

- o Plasma Spray
- o Chemical Vapor Deposition
- o Explosive Bonding.

PLASMA SPRAY

In the plasma spray process, an inert gas, excited by an electric arc to a temperature in excess of 15,000 C, vaporizes the powdered coating material and carries it to the substrate. In this travel from the spray gun the vapor

coalesces into tiny liquid droplets which flatten on impact and rapidly cool. The resultant coating thus has a porous lamellar structure. Porosity can be easily controlled in the coating; densities of 90 percent of theoretical are readily obtained, and up to 98 percent is possible. Oxidation is prevented by means of a skirting stream of the inert gas which completely surrounds the plasma jet.

Advantages of the plasma spray method, compared to the two alternative techniques, include:

- o Allows thick deposits on large, irregularly shaped surfaces.
- o Permits in-situ coating or repairs.
- o Can be scaled up to the referenced ICF structural configuration and dimensions.
- o Experience with a wide variety of coatings (including tantalum) and substrates shows good thermal fatigue and shock resistance.
- o Can be used to deposit other high temperature refractory materials (metals or ceramics).

CHEMICAL VAPOR DEPOSITION

For the chemical vapor deposition of tantalum, TaCl_5 is reduced with hydrogen or zinc in a large retort containing the substrate materials according to one of the following reactions:

These reactions have been successfully used to coat small samples with coating thicknesses up to some tens of microns. The coatings have displayed good

thermal shock characteristics on substrates of Al, Cu, SS316 and Inconel 600. However, this technique would not be useful for applying coating thicknesses up to 1 mm on large samples or for in-situ application.

EXPLOSIVE BONDING

Explosive bonding has been used successfully with a variety of coatings and substrates, and in the range of the required thicknesses. However, the coating would have to be applied prior to forming the coolant tubes, leading to fabrication difficulties. It is therefore not clear, without further study, whether this method can be extended to the ICF configuration.

For the reasons given above, the plasma spray method was judged to be the most attractive of the options considered.

6.3 Verification of Manufacturability

A sheet of HT9 measuring approximately 12" x 4" x 1/8", and part of a 14,750 kg heat poured for the National Fusion Ferritic Steel Program, was provided by the General Atomic Company. The chemical composition of the heat, compared with nominal HT9 values, is given in Table 6-1. Also listed is the composition of the stainless steel 410 (SS410) from which sheets, having dimensions identical to the HT9, were procured. These sheets are being used as dummy samples for confirming procedures used in each step of the process for fabrication of the tantalum coated tubes. Stainless steel 410 was selected because of its availability and similarity to HT9.

Several tantalum metal powders were investigated. Their properties are given in Table 6-2 below. The choice of the powders was based on how well the plasma sprayed and on their oxygen content.

The fine (-325) #3 powder was eliminated for the above reasons. Purer material was significantly more expensive, i.e., price of 600 ppm $O_2 \simeq 3 \times$ price of 1000 ppm O_2 . According to Klopp et al. (1958) in Tietz and Wilson⁽¹⁾ however there is significant change in the mechanical properties

TABLE 6-1
CHEMICAL COMPOSITION OF STEEL SAMPLES

<u>Steel</u>	<u>Chemical Composition (w%, Balance Fe)</u>							
	<u>Cr</u>	<u>Mo</u>	<u>V</u>	<u>Mn</u>	<u>C</u>	<u>Si</u>	<u>Ni</u>	<u>W</u>
HT9 (Nominal)	11.5	1.0	0.3	0.55	0.20	0.4	0.5	0.5
HT9 (Sample)	11.6	1.0	0.3	0.61	0.20	0.24	0.54	0.61
SS410	11.7	0.07	*	0.34	0.13	0.56	0.26	*

*None reported

TABLE 6-2
CHARACTERISTICS OF DIFFERENT TANTALUM POWDERS

	<u>(Kawecki) #1</u>	<u>(Kennametal) #2</u>	<u>(Kennametal) #3</u>
Metal content	NA	99.7	99.7
Gas content			
O ₂ (ppm by wt.)	1175 (2)*	1006 (3)	3745 (2)
N ₂ (ppm by wt.)	75	86	130
Grain Size (mesh)	-100 +200	-200 +325	-325
Spray characteristics	good	fair-good	bad

*Number of determinations averaged

of Ta at room temperature with the increase in oxygen content from 600 ppm to 1200 ppm. The strength and modulus increase significantly while the malleability decreases substantially.

PLASMA SPRAYING PROCEDURE

The plasma spray process has a large number of variables that may be controlled to vary coating quality (density, microstructure, etc.) and adherence, including:

- o Cover gas atmosphere
- o Spray gun - sample distance
- o Powder purity and morphology

In actual application, the coating would be applied inside of the ICF chamber. To simulate these conditions, a rectangular container with a perforated copper tube around the open side was fabricated to provide a curtain of inert argon gas across the open side and over the tube surface. This was in addition to the argon skirt gas of the plasma spray gun itself. Distance and spray pattern were established with tests on the SS410 dummy tubes and extensive experience from other (primarily refractory oxide) coating materials.

The #1 (-100 +200) powder and the #2 (-200 +325) powder were each sprayed on one 12 inch hemi-cylindrical HT-9 substrate using a Plasmadyne plasma spray unit with SG-100 gun. The plasma spray gun itself was equipped with a shield gas attachment which surrounded the plasma spray jet with an argon envelope.

The carrier gas and the arc gas were also both argon. The arc gas was doped with enough hydrogen gas to raise the arc voltage about 10 volts. This was done to increase reducing conditions in the plasma in order to reduce the oxygen content of the sprayed coating as well as improve the particle to particle bonding.

Spraying was carried out as follows. The sample was positioned so that the upper or lower half of the cylindrical surface could be sprayed. The plasma spray gun was set to run at currents, voltages, and gas flow rates indicated in Table 6-3.

At these settings the spraying was performed with the gun held ~3 inches from the substrate (stainless steel) and the spray was swept both parallel and perpendicular to the axis of the cylinder. Spraying was continued for several minutes at a time and stopped when the substrate began to heat up. There was enough fugitive oxygen in the present system to cause some oxidation of the sprayed tantalum which was noticeable when the substrate heated up to a few hundred degrees centigrade. After the substrate had cooled again, the process was repeated until the desired thickness was built up. Typically 5-10 mills were added per spraying using the coarser powder (#1) and 2-4 mills using the finer (#2).

6.4 General Appearance

The surface of the plasma sprayed tantalum has an overall matte finish. On close inspection, small, shiny metallic areas are evident. These are the surfaces of the last droplets to splat against the sprayed surface. Other miscellaneous observations indicate that the plasma sprayed layer is both hard and brittle. Striking the layer on an edge will not indent the layer but cause pieces to chip off. Significantly, the breaks always occur through the bulk of the layer and never at the layer-stainless interface. A steel file will mark the surface of the layer with difficulty and when drilled from the stainless through to the plasma sprayed layer, the tantalum tends to spall off around the hole, but again not breaking at the tantalum-stainless interface.

6.5 Conclusion

The plasma spray method satisfies the requirements established for the manufacture of Ta-steel composites. It can be applied rapidly in-situ and, by proper control of the cover gas during spraying, can produce reproducible

TABLE 6-3
OPERATIONAL PARAMETERS FOR PLASMA SPRAYING Ta

Gun Voltage	40V with H ₂
Gun Current	800 mA
Distance to Substrate	~3 inch
Shield Gas Flowrate	~50 cfh
Arc Gas Flowrate	50 cfh
Carrier Gas Flowrate	15 cfh
Substrate Flush Gas Flowrate	~50 cfh

coatings with good appearance and adherence. In Section 7.0 tests of the strength of the Ta-steel bond under conditions simulating startup and shutdown are described.

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7.0 EXPERIMENTAL STUDY OF INTEGRITY OF Ta-STEEL INTERFACE

The bond between the tantalum coating and the HT9 steel tubes (which comprise the first wall structure) must withstand both the rapid cycling temperature variations from the pellet explosions as well as the stresses resulting from the much longer duration temperature changes experienced during plant startup and shutdown. The test program reported here provides experimental information on the latter concern. Details of sample preparation were given in Section 6.0 and are repeated here for convenience.

7.1 Materials

Tantalum is more difficult to spray than most metals and oxides because of its extreme reactivity to atmospheric gases such as oxygen and nitrogen. This is particularly true at the very high temperatures of the spray particles. To combat contamination by the atmosphere the plasma spray stream and the substrate were both shielded by streams of pure argon (<5 ppm O_2) as described later. This difficulty is accentuated by the fact that available tantalum powders are already contaminated by oxygen, also described later. Several tantalum metal powders were investigated as described in Section 6.0. The choice of the powders was based on how well the plasma sprayed, on their oxygen content, and on their cost. In spite of these difficulties, good looking coatings were achieved on HT9 stainless steel, even though they were embrittled because of oxygen, nitrogen and hydrogen contamination.

The substrate metal was in the form of two hemi-cylindrical sheets of HT9 steel 12 inches long with an inside radius of curvature of $1 \frac{5}{16}$ inches and a thickness of 70 mils. After it was formed and annealed, the exterior radius was sand blasted to clean it and give it a matte finish so the plasma sprayed metal would adhere well.

7.2 Plasma Spraying Procedure

The #1 (-100 +200) powder and the #2 (-200 +325) powder were each sprayed on one 12 inch hemi-cylindrical HT9 substrate using a Plasmadyne plasma spray

unit with SG-100 gun. Figure 7-1 shows the hemi-cylinders before and after being sprayed. The plasma spray gun itself was equipped with a shield gas attachment which surrounded the plasma spray jet with an argon envelope.

The carrier gas and the arc gas were also both argon. The arc gas was doped with enough hydrogen gas to raise the arc voltage about 10 volts. This was done to increase reducing conditions in the plasma in order to reduce the oxygen content of the sprayed coating as well as to improve the particle to particle bonding.

The spraying was performed with the gun held ~3 inches from the substrate (stainless steel) and the spray was swept both parallel and perpendicular to the axis of the cylinder. Spraying was continued for several minutes at a time and stopped when the substrate began to heat up. There was enough fugitive oxygen in the present system to cause some oxidation of the sprayed tantalum which was noticeable when the substrate heated up to a few hundred degrees centigrade. After the substrate had cooled again, the process was repeated until the desired thickness was built up. Typically 5-10 mills were added per spraying using the coarser powder (#1) and 2-4 mills using the finer (#2).

7.3 Thermal Cycle Test Procedure

The thermal cycle test was designed to demonstrate the mechanical stability of the plasma sprayed tantalum coating on HT-9 stainless steel when cycled slowly ($\Delta T \leq 20^\circ\text{C}/\text{min}$) between room temperature and 500°C . This cycle produced thermal stresses in excess of those expected during the startup and shutdown of the ICF power plant.

Two 12 inch pieces were each cut into four 3 inch sections, then drilled and restraining bolts installed so that each half cylinder would not be able to warp during subsequent thermal cycling. They were encapsulated in a pyrex envelope under high purity argon gas ($\sim 2/3$ atm at room temperature) to

Figure 7-1. Stainless Steel Hemicylinders Prepared for Plasma Spraying (Bottom) and with a 40 mil Plasma Sprayed Tantalum Coating (Top)

8158-1

prevent oxidation of the tantalum at elevated temperature in the furnace. Tubulations at the top and bottom were used to flush the samples with high purity argon during encapsulation. See Figure 7-2.

The encapsulated test pieces were loaded onto an elevator platform which was periodically lowered into and raised out of the hot zone of the furnace below. Stainless steel plates acted as baffles to help control convection currents around the test pieces. The temperature was monitored by a chromel-alumel thermocouple inserted from the top which indicated the temperature at the mid-point of the samples. A dummy sample (two pieces of stainless steel in an open glass cyclinder) was installed to replace actual test samples as they were removed. This was done to maintain a similar thermal regime throughout the test.

By running the elevator at 1 inch per minute, the time-temperature profile shown in Figure 7-3 was generated. In the "up" position the samples were cooled further by a small fan blowing on them. The average high and low temperatures were $500 \pm 15^{\circ}\text{C}$ and $35 \pm 5^{\circ}\text{C}$. In total there were four sets of two samples; each set consisted of one sample sprayed with #1 (-100 +200) Ta and one sample sprayed with #2 (-200 +325) Ta. The four sets experienced 0, 20, 50, and 100 cycles respectively.

7.4 Post-Test Analyses

After thermal cycling, the samples were removed from the glass capsules and cut up to make specimens for metallographic evaluation. Specimens were also prepared from the 0 cycle and 100 cycle samples for density measurements, x-ray diffraction, and scanning electron microscope evaluation, and tensile and shear strength measurements.

Figure 7-2. Encapsulated Test Specimens for the Thermal Cycle Test. A Coarse and a Fine Grained Coating were Included in Each Envelope

Figure 7-3. Representative Time-Temperature Profile for the Thermal Cycle Test

8158-4

7.4.1 Density

The density was estimated for coarse and fine grained coatings before and after cycling by weighing small rectangular pieces of the coating for which the volume had been determined. The results indicate that the density of the plasma sprayed coating is from 85 to 91% of the theoretical value of 16.6 g/cc.

7.4.2 Shear Strength

This property was measured to evaluate the strength of the bond between the plasma sprayed coating and the stainless steel substrate as well as the bulk strength of the material itself. To make the measurement, small rectangular pieces were subjected to a shear stress using the device illustrated in Figure 7-4. Pressure was increased on the plunger until the test piece failed. The pressure at failure allowed the shear strength values plotted in Figure 7-5 to be computed. The tests were run so that the shear failure could occur at the interface between the stainless steel and the tantalum if that was the weak plane; however the failure always occurred within the plasma sprayed layer. Hence the values presented in Figure 7-5 are a lower bound for the shear strength of the bond between the two materials.

The source of the scatter displayed by much of the data is not known but is believed to be due to variations in the coating accentuated by the small cross sectional area of the test pieces. Visual observations of the failure surfaces for both 0-thermal cycle and 100-cycle specimens suggested that the plane of failure was between individual flattened droplets of tantalum metal. Significantly, this failure surface for the 100-cycle sample was much freer of tarnish (oxide) than the 0-cycle samples and may be related to the somewhat higher strength exhibited by them.

7.4.3 Tensile Strength

Tensile strength measurements were performed at a strain rate of .05 inch/minute on specimens carefully machined to the configuration shown in Figure 7-6. However, even with careful machining, it was found to be very

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Figure 7-4. Jig to Hold Sample for Shear Strength Measurement

Figure 7-5. Observed Values of the Shear Strength of the Plasma Sprayed Tantalum Coating Parallel to the Interface Between the Coating and the Stainless Steel Substrate for as Formed Samples and Samples Which Have Experienced 100 Thermal Cycles

8158-5

**Figure 7-6. Plasma Sprayed Coating Sample Prepared for Tensile Strength Measurement
Sample is 3" Long**

difficult to keep from breaking the specimens. It was finally necessary to "repair" specimens that weren't too badly broken with epoxy and measure their tensile strength. If the epoxy joint failed, a minimum strength value had been determined; if not, then a legitimate tensile strength was measured. Two out of four specimens could be measured and gave the following results:

<u>Mesh Size</u>	<u>Number of Cycles</u>	<u>Tensile Strength</u>	<u>Comments</u>
-100, +200	0 cycles	~3200 psi	epoxy joint failed
-100, +200	100 cycles	3200 psi	plasma sprayed layer failed

The break, whether due to tension or a bending moment, appeared the same. The broken surface was very rough and, at 50X magnification, layering was evident. Considerable evidence of an intercalated texture was observed indicating that much of the failure had occurred along droplet boundaries.

7.4.4 Metallography

Each grain of tantalum powder is melted in the plasma arc to form a molten droplet which flattens into a pancake as it impacts the substrate. On impact, the melt is quenched. Figure 7-7 shows that the plasma sprayed layer is built up of these "splats" of metal against the cool substrate.

Porosity in the plasma sprayed material appears to be concentrated at the junction between "stacks" of flattened droplets. In the "stacks", the material is very close to theoretical density. At the junctions, "bridging" occurs forming pores.

Figure 7-8 shows that the plane of failure of a shear test specimen of the plasma sprayed layer occurred at the boundaries of the flattened droplets. This observation is consistent with the tarnished appearance of the plane of failure. The tarnish is due to oxidation which would form on the surface of the flattened droplet. Both finished and etched polished sections show variations in color or texture that suggest some contamination of the tantalum

Figure 7-7. An Etched, Polished Section of a Plasma Sprayed Tantalum Layer Produced using –100, +200 Mesh Tantalum Metal Powder (200x)

8158-7

Figure 7-8. Reflected Light Micrograph of a Polished and Etched Section Showing the Plane of Failure of a Shear Strength Test Specimen. This is the $-200, +325$ Material, 0-Cycles (500x)

8158-8

layer. In the next section the possible identification of these contaminants is made from the results of x-ray diffraction analysis and scanning electron microscope (SEM).

7.4.5 X-Ray Diffraction and Scanning Electron Microscope Results

X-ray diffraction analysis was performed on "as sprayed" and "100 cycle" samples of both the fine and coarse coatings. Both Ta_2O_5 and a nitride phase, $TaN_{0.1}$ ⁽¹⁾, were detected in addition to the main component, Ta metal, as indicated in the table below.

<u>Mesh Size</u>	<u>0-cycles</u>	<u>100-cycles</u>
-100, +200	tr Ta_2O_5	tr Ta_2O_5 , minor $TaN_{0.1}$
-200, +325	tr Ta_2O_5	tr Ta_2O_5 , minor $TaN_{0.1}$

Scanning electron micrographs (Figures 7-9 and 7-10) show a layered texture like that already observed in optical micrographs. Light and dark regions were analyzed and only tantalum was detected. At the same time the count rate for tantalum was observed to be about 20% higher in the lightest areas than in the darkest. Since the SEM system does not detect elements with an atomic number less than sodium, any compounds containing oxygen, nitrogen, carbon, hydrogen, etc. would only indicate tantalum when analyzed. These results are consistent with the x-ray data and suggest that oxide, nitride, or similar compounds of tantalum are distributed throughout the plasma sprayed layer.

There appear to be at least two textural differences between the "as sprayed" and the "100 cycle" coatings made from both tantalum powders. First, the thermal cycled coating seems to have etched more pronouncedly along the boundary between individual flattened droplets. Second, there seems to be more contrast between light and dark areas in the cycled specimens. Both of these observations suggest that some compound segregation occurred during thermal cycling; this is consistent with the formation of the nitride detected by x-ray diffraction.

Figure 7-9. SEM Micrographs of –200, +325 Coating After 100 Thermal Cycles (Top) and as Formed Bottom. More Dark Areas are Observed in 100 Cycle Sample. SEM-EDAX of Both Light and Dark Shows Only Ta but a Comparison of the Count Rates Suggest that the Light Areas are ~20% Richer in Ta. Oxygen or Nitrogen would not be Detected. Both Samples Etched. The Interface with the Substrate is to the Left.

Figure 7-10. SEM Micrographs of –100, +200 Coating after 100 Cycles Top and as Formed (Bottom). Etching has Attacked Boundaries Between the Flattened Droplets

7.5 Discussion of Results

A one mm thick tantalum metal coating was produced via plasma spraying using readily available tantalum metal powder. This coating was not degraded by thermal cycling to 500°C under an argon atmosphere and had mechanical strength levels more than adequate to support itself reliably.

Our evaluation of the coating both by visual and instrumental analysis suggested that other phases were present. The fact that only tantalum was detected by the scanning electron microscope indicates that only elements lighter than sodium could be present in addition to tantalum, a fact consistent with the X-ray diffraction analyses results, which indicated the presence of an oxide and a nitride. The other element probably present is hydrogen which is readily soluble in tantalum at red heat and could have been picked up from the arc gas during plasma spraying. Since tantalum forms a variety of nitrides and oxynitrides⁽²⁾ in addition to oxides and hydrides,⁽³⁾ including limited solid solutions with these elements, it is not possible to sort out all the possibilities with the limited analytical data available. However, all of the data are consistent with these types of compounds producing the observed heterogeneity.

The brittleness of the tantalum coating mentioned earlier is certainly the result of chemical contamination by oxygen, nitrogen and hydrogen. Since one of the attractive properties of tantalum metal as a first surface coating is its malleability, an effort should be made to reduce the level of gas (O, N, H) contamination so that a malleable coating is produced. According to Reference (1) tantalum is quite malleable if its oxygen content is below 400-600 ppm. The present coating has about twice that much oxygen in the starting material and an unknown amount from the plasma spraying procedure. The final amounts of hydrogen and nitrogen are also unknown. The potential sources of contamination are known but their relative importance is not known at this time.

To identify the sources of gas contamination in our study, the gas content of both tantalum metal coatings, "as-sprayed" and after 100 thermal cycles, should be measured. The first measurement would indicate the level of

contamination that occurred during the plasma spraying. The second would indicate whether additional contamination occurred during encapsulation and thermal cycling and may explain the appearance of the $\text{TaN}_{0.1}$ phase identified by X-ray diffraction analysis. These analyses would indicate at what stage of processing and to what degree contamination by oxygen, nitrogen, and hydrogen had occurred.

In the next phase of experiments, the tantalum coating will have to withstand thermal shocks of $\sim 2000^\circ\text{C}$ ($500^\circ\text{C} \rightarrow 2500^\circ\text{C}$) from nanosecond pulses at 10 per second to simulate its ultimate application. Since the present coating is quite brittle due to its oxygen content, its ability to withstand that level of thermal shocking is unknown. Low oxygen content tantalum powder could be made in the lab by hydrogen embrittling malleable tantalum wire, grinding to a suitable grain size and vacuum extracting the hydrogen. The tantalum powder would be sprayed and vacuum baked to remove any hydrogen remaining in the coating. This should produce a highly pure, malleable tantalum coating suitable for the appropriate engineering tests, with a much higher probability of success. Another possibility would be to use tantalum wire as a feedstock to the plasma torch.

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8.0 SCOPING DESIGN OF EXPERIMENT TO STUDY TRANSIENT EFFECTS IN Ta COATING

INTRODUCTION

The basic problem in fusion reactors is the design of the first wall of the reactor chamber. This wall must withstand the repeated onslaught of the combined effects of radiation and ions produced by the pellet explosions. Recently Westinghouse proposed a dry first wall comprised of 0.18 cm wall steel tubes coated with approximately 0.1 cm of tantalum.⁽¹⁾ The tantalum coating resists the effects of the radiation and ion bombardment, while liquid lithium metal, flowing within the tubes, carries away the heat produced.

Any wall protection scheme must last for years in the very hostile environment of the ICF reactor. Although many analytical studies have been performed on these schemes, many uncertainties remain which can only be resolved by experimental measurements. These uncertainties include, but are not limited to, the effects of:

- o Very short but intense temperature spikes
- o Sputtering
- o Chemical reactions
- o Fatigue

Another feature of interest in laser-driven ICF reactors is an examination of the effects resulting from radiation trapping and blast waves generated in the 0.1 Torr of gas present in these reactors. The latter effects will also occur in light ion-driven reactors which require a high pressure gas for their operation, but will be absent in heavy ion-driven reactors which require gas pressures below 10^{-4} Torr.

To determine the validity of the dry wall design as well as other wall protection schemes, a simulation facility must therefore be built to investigate the net effect of these complicated processes. To facilitate the eventual construction of such a facility, a scoping study has been undertaken

and is reported herein. The next section outlines the general system considerations for this facility, and is followed by sections on the pulse sources and experimental diagnostics.

8.1 System Considerations

8.1.1 Sample Size

In a well designed ICF spherical reactor chamber the X-ray and ion fluxes will be normal to the wall. Lateral temperature gradients along the wall surface should therefore not be significant and must be avoided in any simulation facility. On the other hand, temperature gradients normal to the surface will be very large and will have significant time dependencies. This behavior must not be significantly modified by the design of the simulation facility.

In the simulator a circular beam of radius a and irradiance H will impinge on a small sample of the first wall. The lateral heat conduction in the sample, relative to the normal heat conduction into the sample, will be a function of the beam radius and sample size. Consider first the case where the sample is much larger than the beam. The heat flow in the lateral direction is difficult to calculate and is beyond the intent of this study. However, the ratio of the lateral heat flow to the normal heat flow is, to zero order, simply the ratio of the areas through which the heat flows:

$$\begin{aligned} f &= 2\pi \lambda / \pi a^2 \\ &= 2\lambda / a \end{aligned} \tag{8.1}$$

where λ is the effective depth of penetration of the heat flow.

The depth λ at the end of the heating pulse can be estimated from the analytical solution for the sudden heating of a semi-infinite solid:⁽²⁾

$$\lambda \approx 0.6 \sqrt{\alpha \tau} \tag{8.2}$$

In this expression the right hand side is the analytical solution for the depth at which the temperature is 1/2 the value at the surface. In this expression τ is the temporal width of the heating pulse and α is the thermal diffusivity of the coating:

$$\alpha = k/\rho C \text{ cm}^2/\text{sec} \quad 8.3$$

where k , ρ , and C are the thermal conductivity, mass density, and specific heat, respectively. For tantalum

$$\alpha = 0.2174 \text{ cm}^2/\text{sec} \quad 8.4$$

The X-ray pulse nominally lasts for tens of microseconds. For a 100 nsec pulse the above expressions indicate that the effective depth at the end of the pulse will be $0.9 \sigma_m$ whereas the effective depth will be $28 \sigma_m$ for a 100 nsec pulse. For a 1% loss factor (Equation 8.1) the beam radius must be 0.55 cm and the sample size must be larger than this, i.e., it must have more than 0.95 cm^2 of area.

These requirements are sufficient if the main concern is intrapulse behavior. However, the relaxation behavior of the wall between pulses is also of interest but will require a much larger sample. For a 10 pps reactor the time between pulses is 100 msec. However, in this case it is not the time which is important but rather the coating thickness and we can simply set the penetration depth to that thickness. Thus, for a 1.0 cm thick coating and a 10% loss factor, the beam radius must be 2 cm and the area of the sample must be larger than 13 cm^2 . This sample size should be no problem to fabricate, but the 2 cm radius beam can be a severe limitation on the pulse sources since the energies required from the sources scale as the beam radius squared.

An alternative to large sample sizes and beam radii is to use a very small sample in which the entire surface is irradiated with the incident beam. In this case the lateral flow is reduced since it occurs by the much less efficient radiation of energy from the sample sides. This approach has many advantages since the thermal behavior of the wall sample can be viewed

directly along the side, and the energy requirements on the beam are much less. The main disadvantages are the difficulties of handling small samples, and correlating the thermal behavior of the small sample to the expected behavior of the large reactor wall.

8.1.2 System Configuration

The general system configuration is shown schematically in Figure 8-1. A pulsed X-ray source of the proper energy and time duration is fired first into a relatively large cylinder which contains a controlled gas environment. The beam travels through the gas and strikes the sample at the opposite end. At the appropriate time a pulsed ion source of the proper energy and time duration is fired into the cylinder and its beam also strikes the sample. The firing of the two beams is controlled so as to simulate the arrival of the X-ray and ion pulses at the first wall of an ICF reactor.

The controlled gas environment may be a near vacuum (10^{-5} Torr) to simulate the heavy ion reactor, may vary up to 10^{-1} Torr to simulate the laser reactor, or up to 100 Torr to simulate the light ion reactor. The gas constituents may also be varied to simulate the actual environment expected in a reactor. The gas handling apparatus for controlling this environment is not shown in Figure 8-1.

For gas pressures above 10^{-2} Torr, pressure waves will be formed in the gas from the energy deposited by the X-ray and ion beams. The effect of these shock waves on the sample may be important. To simulate these shock waves, and the accompanying gas heating, the radius of the muffler should only be slightly larger than the radii of the beams. The muffler, a tube containing numerous holes, damps the shock waves traveling toward the tube walls. By preventing these waves from striking the sample after being reflected off the wall, the simulation of a large reactor cavity which would not have these walls is not degraded. Acoustical absorbing materials may also be placed between the muffler and the outer wall.

The length of the tube is unimportant for pressures below 10^{-2} Torr. However, for higher pressures the tube length becomes important and in reality

Fig. 8.1—General system configuration for simulation facility

should be the same as the radius of the ICF reactor cavity being simulated. Although many gas pressure effects scale as the product of pressure and distance, thereby allowing the tube length to be shortened in exchange for a higher gas pressure, such scaling is not universally valid. In fact, many nonlinear effects such as radiation trapping may be significant.⁽³⁻⁵⁾

Since sputtering of the sample may be important, it also would be desirable to heat the entire tube to a temperature higher than the steady-state temperature of the sample. This would allow any sputtered material to recondense onto the sample rather than on to the tube walls.

Another feature which should be incorporated into the simulator is the ability to turn the sample so the beams may strike its surface at an angle. This capability would allow the simulation of cylindrical reactor cavities as well as spherical cavities.

Also sketched in Figure 8-1 is apparatus for monitoring the performance of the beams and for studying the behavior of the sample. Not shown is an apparatus for monitoring the state of the controlled gas environment. The apparatus for monitoring the beams and gas are rather standard and will not be discussed further. However, the pulsed energy sources and sample diagnostic methods are not so conventional and will be discussed in the next two sections.

8.2 Pulsed Power Sources

8.2.1 X-Ray Sources

The X-ray spectrum from the pellet explosion can be divided into two parts: hard X-rays (10 keV or above) and soft X-rays (1 keV or lower). The hard X-rays penetrate deeply into the chamber wall and heat slightly a large volume of the wall. Conversely, the soft X-rays have little penetration and generate a large temperature spike at the surface of the wall. Although the hard X-rays can be safely neglected, it is important to simulate the soft X-ray pulse, which only lasts for tens of nanoseconds and produces an irradiance of 10 kW/cm^2 or higher.

One source of X-rays is the synchrotron radiation from an electron accelerator which constrains the electrons in a curved path. This radiation is tunable with a broad and intense energy spectrum. However, it is only generated in large, cumbersome devices operating at high energies and cannot be considered seriously for the ICF simulator facility.^(6,7)

The simplest and most promising X-ray source for the simulator facility is one which uses the electron impact on metals to produce the X-rays. The electrons are obtained from a cathode by either field emission and/or thermionic emission. The electrons are then accelerated and focused onto the target by electromagnetic means which can be easily pulsed. The basic design of these compact X-ray tubes has been described by Barrie.⁽⁸⁾

When an electron enters a metal target, three types of radiation are produced:

- o Characteristic
- o Bremsstrahlung
- o Transition

The X-ray energies of the characteristic radiation spectrum are quite narrow and intense since they arise from the electronic transitions of the atoms in the target. Since the spectrum of the X-ray source should fairly closely match the expected ICF spectrum, it is important that the electron target material be, as much as is possible, the same material as the ICF pellet, i.e., tantalum.

Bremsstrahlung radiation is a volume effect resulting from multiple collision processes as the electrons lose their energy and are finally brought to rest. This is the most common source of X-ray radiation and is very promising for the simulator facility. Hughes, Heumier, and Griffin have even published the Bremsstrahlung and characteristic radiation spectrum of tantalum under bombardment with 3 keV electrons.⁽⁹⁾ To obtain very soft X-rays, where absorption by the target may be a serious problem, the solid metal target may be replaced by a metallic vapor obtained by evaporating a slug of solid metal.⁽¹⁰⁾

In contrast to bremsstrahlung radiation, transition radiation is a surface effect which occurs when a charged particle crosses the boundary between two media with different dielectric constants. Since it is a surface effect, the radiation from multiple boundaries in a periodic structure can be made to add coherently in certain directions. As a result very intense and directional X-rays are obtained which may be two orders of magnitude higher than those obtained from a synchrotron source.⁽¹¹⁾

Bremsstrahlung, in general, is a relatively strong source of radiation at the short wavelengths whereas transition radiation is strong at the long wavelengths. The two forms of radiation therefore complement each other. This complementary nature is seen in the spectra from tantalum, where the bremsstrahlung radiation decreases from 100 Å to 1000 Å beyond which it is very weak. By contrast, the transition radiation decreases from 500 Å to 5000 Å beyond which it becomes very weak.⁽¹¹⁾ The main limitation of these X-ray sources is the electron gun which, instead of generating X-rays, can also be used to heat the wall sample directly. Because of the small penetration depth of soft X-rays, heating by electrons is quite a good approach. The two major problems with the electron simulation technique are that direct electron heating cannot simulate the characteristic radiation, which may or may not be significant, and that the generation of shock waves in gases will be quite different for electron beams than for X-ray beams.

Although commercial electron guns satisfying the performance specifications needed for the simulation facility do not exist, there are some that come close and may be modified for use in the facility. For example, a transverse electron gun design developed at the Colorado State University can give 80 kW in a 2 μs pulse of 1-10 kV electrons.⁽¹²⁾ Westinghouse has a wire ion plasma (WIP) 54 kW, 150 kV electron gun capable of operating at 10 kHz with pulse widths of 200-600 nsec.

It is clear that some development work remains to be completed before an electron gun with the proper specifications can be used either for directly heating the sample or for generating the desired X-ray beam. A more reasonable approach, at least in the short term, may be to use laser irradiation to heat the sample. Commercially available lasers already exist

which are satisfactory, such as the hydrogen fluoride or deuterium fluoride chemical laser (50-200 nsec pulses, 2-600 mJ/pulse, 0.1-2 cm beam radius) and the carbon dioxide laser (50 nsec - 75 μ sec, 0.03-75 J/pulse, 0.2-5 cm beam radius).⁽¹³⁾

In addition to the problem of the high reflectivity of metallic surfaces, the main problem with using lasers is in the fact that interaction of the laser radiation with the sample surface is complicated and undoubtedly does not simulate too closely the X-ray radiation effects.^(14,15) On the other hand, if the effects are primarily caused by simple heating and heat flow, laser irradiation should be quite satisfactory. In any event, lasers are presently the best candidate for the "X-ray pulsed source."

8.2.2 Ion Sources

The arrival of the ion pulse at the ICF reactor wall occurs many microseconds after the X-ray pulse, and lasts for tens of microseconds with an irradiance of 50 kW/cm² or higher. Although the ion pulse has much more energy, its significantly lower power reduces the heating effects in the chamber wall. As with the X-ray pulse, the ions have relatively little penetration into the surface, indicating that the thermal effect of an ion pulse can be closely simulated by a surface heating pulse such as can be obtained with a laser beam. However, the long term effects of sputtering and ion contamination of the sample can only be studied with a true ion beam comprised of ions that closely approximate the ions expected from an ICF pellet explosion.

There are many ionization techniques and concepts used to generate ion beams. Unfortunately, the high ion current (>4 A/cm²) required for the ICF ion pulse simulator limits the possibilities to only two:

- o Field Ionization Source
- o Spark Source

Both sources, however, produce small radius beams.

The field ionization source obtains its ions by ionizing atoms near a sharp point where the electric field is extremely high, whereas the spark source generates its ions in a spark discharge, usually between two electrodes fabricated of the atoms from which the desired ions are to be obtained. The very nature of these processes restricts the field ionization source to relatively low ion currents ($<10^{-8}$ A). On the other hand, large area spark sources ($5-7 \text{ cm}^2$) have produced 50-200A of light ions in lasting for seconds pulses.⁽¹⁶⁾ It thus appears that an improved spark source would be capable of producing the desired ion beam.

8.3 Experimental Diagnostics

8.3.1 Measurements Desired

The effects of a thermonuclear fusion lead to the production of soft x-radiation and rapidly travelling ions. The X-ray pulse produced by a single pellet is expected to last some tens of picoseconds, and, for a 10 meter radius chamber, will reach the wall about 35 nsec after the implosion. The ion pulse will arrive approximately $1 \mu\text{s}$ later, and is expected to last many microseconds. The primary affect of the two pulses is to cause very rapid increase in the surface temperature of the wall, with the maximum temperature, produced by the soft X-rays reaching as high as perhaps 3000K, close to the melting point of tantalum. Such a rapid rise in temperature will produce a momentary deformation of the surface and an acoustic wave that will propagate both along the surface and into the wall. These effects, in turn, will produce stresses in the chamber wall that will effect the wall integrity.

In studying the immediate effects of a simulated pellet explosion, a primary measurement must be the temporal and spatial dependence of the temperature. Secondary desired measurements are those that record the surface deformation, and the propagation of the acoustic wave produced.

The measurements ideally should be taken with a time resolution equal to or less than the fastest significant event, i.e., $t < 10^{-6}$ sec, by methods that do not perturb the phenomena to be measured, and in an environment that conservatively can be labeled as hostile. Although mathematical modelling of

the effects of a pellet explosion show that the temperature rise and decay is confined to the Ta coating,⁽¹⁾ clearly some effects will be felt by the steel substrate. Thus, measurements across the entire cross section of the wall are desired, over a time period of perhaps milliseconds, considerably longer than merely the duration of the pulses. The ultimate aim is to acquire sufficient data to permit calculations of the stresses produced. Long term effects on the vessel wall, such as bulk damage, effects on the Ta to steel adhesion, etc., that result from repeated exposure to pellet fusions can be studied by more conventional methods.

In the sections that follow, we describe possible measurement techniques for obtaining the desired information. Following this, we discuss the facilities that are available for these measurements, and the possible difficulties to be encountered.

8.3.2 Temperature Measurement

GENERAL DESCRIPTION

As mentioned above, the temporal and spatial dependence of the wall temperature following a pellet explosion constitutes a basic measurement. The two most common methods of temperature measurement are use of a thermal probe, such as a thermocouple, and optical pyrometry. Thermocouples are inherently slow, and have the further disadvantage of having to be attached to the surface. The attachment perturbs the surface and may distort the results.

Optical pyrometers, on the other hand, are used remotely, and are therefore nonperturbing. Since they measure some property of a radiating surface, they are also ideal for measurements at high temperatures, where the radiation intensity is high. The commonly used optical pyrometers are intensity pyrometers--they compare the intensity of the surface radiation within a narrow wavelength range with the intensity of an adjustable internally heated source. This usual method of measurement is also very slow, however, and is not in its conventional form capable of following temperature changes of

microsecond durations. Intensity pyrometry is also more subject to inherent errors, such as window transmission and the deviation of the source of radiation from that of a black body, than the method about to be described.

The method proposed here is two-color radiation pyrometry. Like conventional pyrometry, it is a remote sensing technique and therefore nonperturbing. Instead of measuring the intensity of the radiation of a surface at one wavelength, it utilizes the ratio of the intensities at two wavelengths, generally close to one another. Because the ratio is being measured rather than an absolute intensity, the method requires no correction for the transmission of the window providing the window has the same transmission at both wavelengths, a requirement that is usually easily met.

TWO-COLOR PYROMETRY

The measurement of temperature by optical pyrometry is based on the fact that for a black body, the power emitted at a given wavelength is determined by the temperature alone, and is given by the well known expression:

$$I_0(\lambda, T) = \frac{2hc^2}{\lambda^5} \frac{1}{\exp(hc/\lambda kT) - 1} \quad 8.5$$

Where c is the velocity of light, λ is the wavelength, and h and k are Planck's and Boltzmann's constants. For real surfaces, which are generally not black bodies, the emissivity, $\epsilon(\lambda, T)$ must be taken into account. Equation (8.5) becomes

$$I(\lambda, T) = \epsilon(\lambda, T) I_0(\lambda, T) \quad 8.6$$

Equation (8.5) forms the basis for intensity pyrometry. For accurate temperature measurements, corrections must be made for the emissivity of the surface. Consequently, there exist many tables of emissivity versus temperature and wavelength for various materials.⁽¹⁷⁾

Two-color radiation pyrometry depends not on a determination of intensities, but on a measurement of the ratio of the relative radiation intensities at two different wavelengths. Let R be the ratio of the radiative intensities at λ_1 and λ_2 . Then, since $\exp(hc/\lambda kT) > 1$,

$$R = \frac{I(\lambda_1)}{I(\lambda_2)} = \left(\frac{\epsilon_1}{\epsilon_2}\right) \left(\frac{\lambda_1}{\lambda_2}\right)^{-5} \exp\left(\frac{hc}{kT}\right) \left(\frac{1}{\lambda_2} - \frac{1}{\lambda_1}\right) \quad 8.7$$

It is worthwhile to compare intensity pyrometry with two-color pyrometry. For intensity pyrometry, a temperature accuracy as high as $\pm 5^\circ\text{K}$ can be obtained at 2000°K with NBS calibrated reference sources. The dependence on an accurate knowledge of the emissivity is not great. For example, at 2000°K , and at $0.65 \mu\text{m}$, a 10% error in the emissivity leads to a 0.65% error in temperature. An error analysis of the two methods indicates that the two-color method can be generally more accurate and more sensitive.⁽¹⁸⁾ In particular, if ϵ_1 and ϵ_2 vary with temperature in the same way, errors in two-color pyrometry can be very small.

SENSITIVITY OF TWO-COLOR PYROMETRY

The sensitivity of two-color pyrometry (assuming constant emissivities) is given by

$$\frac{dR}{dT} = \left(\frac{\epsilon_1}{\epsilon_2}\right) \frac{hc}{kT^2} \left(\frac{1}{\lambda_2} - \frac{1}{\lambda_1}\right) \quad 8.8$$

dR/dT clearly depends on the choice of wavelengths. The closer the wavelengths are to one another, the lower the sensitivity. Two convenient wavelengths for which emissivity data is available are $\lambda_1 = 0.665 \mu\text{m}$ and $\lambda_2 = 0.463 \mu\text{m}$ ⁽¹⁷⁾. Approximate values of ϵ_1 and ϵ_2 are shown in Figure 8-2. For these wavelengths,

$$R = 0.1636 \left(\frac{\epsilon_1}{\epsilon_2}\right) \exp\left(\frac{9.45 \times 10^3}{T}\right), \text{ and} \quad 8.9$$

Figure 8-2. Emissivity Vs. Temperature for Ta

$$\frac{dR}{dT} = \left(\frac{\epsilon_1}{\epsilon_2} \right) \frac{9.45 \times 10^3}{T^2} R \quad 8.10$$

Values for ϵ_1 , ϵ_2 , R , and dR/dT are given in Table 8-1 for temperatures of interest.

Table 8-1 indicates that, to cover the entire temperature range in a single measurement, a dynamic range of $1818/3.48 \sim 500$ is required of the detector without a change in sensitivity. With a detector accuracy of 1%, a 10 degree change at 3000K can be determined. This is quite a bit better than is needed. The dynamic range is a function of the wavelengths chosen and can be made smaller or greater by choosing wavelengths that are closer or further apart.

IMPLEMENTATION

To obtain the temperature profiles in the necessary times requires a detector with the following properties:

- o A speed of response of $<10^{-6}$ sec;
- o A dynamic range of ~ 1000 or greater;
- o Adequate sensitivity;
- o Ability to obtain spatial information.

Photomultipliers are generally capable of easily meeting the first three requirements. The fourth requirement is best met by using a vidicon detector. A proposed method of measurement utilizing a vidicon detector is shown in Figure 8-3.

Imagine surface bombardment with a very short burst of energy simulating a pellet explosion. The temperature will instantaneously reach a very high value within the irradiation zone. At some later time, t , as heat diffuses both into and along the surface, the temperature of the surrounding area will have increased, while the irradiation zone temperature will have diminished. An image of this surface on an area-sensitive detector will produce a signal

TABLE 8-1
VALUES OF EMISSIVITY (ϵ_1 and ϵ_2), RATIO OF
RADIATIVE INTENSITIES AT λ_1 AND λ_2 (R), AND
SENSITIVITY OF TWO-COLOR PYROMETRY (dR/dT) AT TEMPERATURES OF INTEREST

T, K	ϵ_1	ϵ_2	R	dR/dT
3000	.388	.426	3.48	3.65×10^{-3}
2500	.399	.444	6.44	9.74×10^{-3}
2000	.414	.464	16.46	3.89×10^{-2}
1500	.432	.489	78.71	3.31×10^{-1}
1000	.452	.517	1818	17.2

Figure 8-3. Schematic Design for High-Speed Two-Color Optical Pyrometry

variation across the face of the detector that is proportional to the radiation emitted by the heated surface over the area being imaged. If the detector is gated so as to be active for a short time Δt and delayed by a time t after the time of the irradiation, the detector signal would correspond to an instantaneous measure of the profile of the surface temperature at time t after irradiation. By taking a series of these temperature "snapshots" with varying delay times, t , it is possible to obtain a series of profiles showing the time development of the diffusion of heat across the surface. By imaging the cross section of the irradiated plate, a similar set of profiles could be obtained for the diffusion of heat into the wall.

The above description does not yet provide the temperature information needed. To obtain the temperature, we split the initial image into two images. These are focussed one above the other on the face of a two section vidicon. Each image is identical except that each passes through a separate narrow band optical filter tuned to $0.665 \mu\text{m}$ and $0.463 \mu\text{m}$ respectively. The point by point ratio of the two filtered signals generated across the vidicon provides the information needed to calculate the temperature profile.

Vidicons that have the above requirements are commercially available. Listed below are the characteristics of an EGG model 1254 SIT vidicon detector:

Target Area:	12.5 mm high x 16 mm wide
Spectral Range:	0.350-0.800 μm
Dynamic Range:	1×10^4
Gating Speed:	4×10^{-8} sec, minimum
Number of Horizontal Channels:	256
Sensitivity:	2 photons/count
Uniformity of Sensitivity:	$\pm 10\%$ over/cm area

The instrument used with the above vidicon, a PAR Optical Multichannel Analyzer, has within it the potential computer power to store the instantaneous images obtained, to obtain the ratios, and to perform the necessary calculations needed to generate the temperatures. Two such instruments with somewhat different characteristics exist in the Westinghouse

laboratories. Thus, the basic capabilities for obtaining the temporal and spatial variation of the temperature following a simulated first wall irradiation already exist in our laboratory.

8.3.3 Measurement of Surface Deformation

GENERAL DISCUSSION

As mentioned in Section 8.2, rapid intense heating of a localized region of a simulated first wall sample will bring about a deformation of the surface. This deformation will be related to the stresses created within the wall. Because the simulation will involve relatively small sample areas, the deformations produced by the heating are expected to be small, despite the very high temperature range involved. (For example, the linear expansion coefficient of Ta at 2000K is $\sim 7.4 \times 10^{-6}$ mm/K). Thus, macroscopic measurements of displacement are not suitable. In addition, the requirements for remote, non-invasive measurement techniques applies equally well to these measurements as they do to temperature, thus eliminating such devices as strain gauges. The preferred method involves the use of optical techniques, and the conventional way of optically measuring small displacements is by interferometry. Conventional interferometry, such as Mach-Zehnder or Michelson, is very difficult to adapt to the present problem. A much more appropriate technique is that of holographic interferometry, and in particular, pulsed holographic interferometry. The latter is particularly suited to the present problem not only because it permits measurements on a scale of 2×10^{-8} sec, but also because it eliminates many of the problems encountered in cw holography.

HOLOGRAPHIC INTERFEROMETRY

A hologram is a recording, usually on a high resolution photographic plate, of the stationary interference pattern produced by the interference of light coming from a reference beam and that produced by the object in question, called the "scene" or "subject" beam. Because the hologram is itself an interference pattern, it is important to differentiate between the

interference pattern that constitutes the hologram, a microscopic interference pattern, and the macroscopic pattern produced by the interferometer which measures the deformations induced in a surface.

To produce a hologram, one needs two coherent beams from a common source, one to serve as a reference beam, and the other to illuminate the object. Both beams originate from a common source. As shown in Figure 8-4, an expanded laser beam passes through a beam splitter. One portion of the beam is the reference beam, which falls directly on a high resolution photographic film. The remainder illuminates the subject, which in this case is the ICF wall surface. The light reflected from the subject falls also on the photographic plate, and constitutes the subject beam.

The microscopic stationary interference pattern recorded on the film constitutes the hologram. To view the hologram, the film is processed and examined with coherent illumination. A coherent source is allowed to illuminate the film along a direction identical to that taken by the original reference beam. An observer at A looking through the film will see a virtual image of the subject in a position identical to that occupied by the original subject. If on the other hand, the observer is at B and views the film by illumination in a direction opposite to that taken by the reference beam, as shown by the dotted lines, he will see a real image where the subject stood. Three dimensional images can be formed by diffusing the subject beam, for example by interspersing a diffuser plate between the beam splitter and the subject.

To form a holographic interferogram requires a double exposure taken on the same film. The first exposure is that of the subject in its normal unperturbed position. The second exposure is identical to the first, but with the subject in its deformed state. The photographic film then contains a record of two holograms, corresponding to the normal and to the deformed subject. When the hologram is processed and viewed as described above, one sees an image of the subject with superimposed light and dark interference fringes. These fringes correspond to phase differences between the two nearly identical images of the undeformed and deformed subject. As in normal interferometry, dark fringes appear at path length differences corresponding

Fig. 8. 4— Schematic arrangement for producing holograms

to integral multiples of $\lambda/2$. By counting fringes, displacements can be determined to a fraction of the wavelength. To form a permanent record, a normal photograph is taken of the holographic image.

Double exposure holographic interferometry has some very important advantages over normal interferometry. Among them are:

- o Since the interference pattern compares only the initial state of a surface with a displaced state, effects due to imperfect windows and other optical distortions cancel out, and need not prevent the interference pattern from forming. Normal interferometry requires the use of optically flat windows and surfaces.
- o For similar reasons, interference holograms of rough or curved surfaces can easily be obtained. Classical interferograms of such surfaces are very difficult or impossible to obtain.
- o As indicated earlier, three dimensional interferometry with large depth of field is possible with diffuse subject illumination. This has no counterpart in normal interferometry.
- o Much greater ease of alignment, a matter of great importance for complex experiments.

PULSED INTERFEROMETRY

For high speed phenomena, pulsed holographic interferometry is required. The basic arrangement is identical to that pictured in Figure 8-4, except that a pulsed laser is used as the coherent light source. There are, however, some practical differences, mostly advantageous. These derive from the fact that during the taking of a hologram the total optical path length must not vary by more than $\lambda/2$. C.w. holograms require considerable overall mechanical stability of the apparatus. Because the exposure times are of the order of 2×10^{-8} sec, pulsed holograms are virtually immune to vibration. As with c.w. holograms, it is important that between the initial hologram and that of the displaced subject, only the desired subject must be displaced or deformed;

otherwise the holographic interferogram will record the total difference in path length due to all contributing motions. It is also desirable that the illumination for the two exposures be nearly equal in order to produce fringes of maximum contrast.

IMPLEMENTATION

The best source is a Q-switched ruby laser. Pulse widths of $\sim 2 \times 10^{-8}$ sec are normally obtained. These speeds are more than adequate for the present diagnosis. Pulse-to-pulse stability of 10% is possible, and is adequate for good contrast. A coherence length of ~ 1 m is desirable and is available with single mode ruby laser oscillators or oscillator-amplifier combinations that are specifically designed for holographic use. High speed high resolution films have been developed in recent years, designed specifically for pulsed ruby laser holography. The literature is replete with examples of pulsed holographic measurements of high speed phenomena.^(19,20) The application of pulsed holographic interferometry should in principle, therefore, present no serious problems in the present application. Indeed, the fact that the wall surfaces will be essentially specular makes the measurements relatively easy to perform.

As with the measurements of temperature, the deformation measurements would consist of obtaining interferograms of the surface at varying times, t , following the simulated pellet explosion. The sequence of events would be as follows:

- o The laser is pulsed, and an exposure is obtained of the unperturbed wall surface;
- o The simulated fusion explosion occurs;
- o At some variable time t following the explosion, the laser is pulsed again and a second exposure is obtained.

The interferogram obtained from 1 and 3 is a record of the deformation present at time t after the pellet explosion. By varying t , a complete history of the deformations can be obtained.

As with temperature measurements, it may be desirable to obtain interference patterns for the edge as well as the Ta surface. It may also be worthwhile observing the deformation of the rear surface of the wall to measure propagation through the wall.

8.3.4 Discussion of Diagnostic Techniques

Much of the success of the diagnostics described above will depend on the design of the overall simulation system. The presence of suitable windows, viewing angles, and adequate space for the measurements will be critical, and will need to be incorporated early in the design of the facility. A major uncertainty is the amount of electrical and radiation shielding that will be necessary. Because both sets of proposed measurements are optical, some immunity from radiation can be gained by choosing longer measuring distances. In the case of the proposed temperature measurements, a critical element is a suitable imaging system for the two-color pyrometry. The use of a vidicon for this purpose is novel. Fortunately, alternative methods are possible for performing high speed two-color pyrometry (for example, with conventional photomultiplier tubes) so that the measurements are not wholly dependent on the exact method described here. Since all of the proposed measurements are sequential in nature, that is to say, measurements are to be taken at varying times, t , after the simulated explosion, it is important to maintain a high degree of stability and reproducibility in the simulated explosions from pulse to pulse. A variation of $\pm 10\%$ is probably adequate. Larger variations may require some sort of averaging of events, which can increase the time required to obtain good results. Finally, while it would be desirable to measure the temperature and surface deformation simultaneously, such simultaneous measurements are likely to present many complications. It will generally be sufficient to obtain the two measurements independently, and to correlate them later. Through such correlations, it should be possible to theoretically deduce the stresses induced in the ICF first wall in actual fusion experiments.

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9.0 EVALUATION OF SOLID LITHIUM BLANKET CONCEPT

One of the contract objectives, prompted by safety and cost considerations, is design of a blanket that uses solid lithium compounds as tritium breeding materials. Three blanket design options were considered initially: 1) liquids other than lithium; 2) stationary solids; and 3) circulating solids. Lead lithium eutectics, specifically $\text{Li}_{17}\text{Pb}_{83}$, were candidates for the liquid blanket, but have several disadvantages: low thermal conductivity, low lithium content, corrosive behavior similar to that of liquid lead, high density, and poorly-known thermal and chemical properties. A static solid blanket requires a relatively complex heat removal system, since helium or water must be circulated throughout the blanket, as well as a system for in-situ removal of tritium. The success of the latter depends on tritium release characteristics, which are uncertain and may be significantly altered by effects of irradiation and cyclic stresses on the microstructure. The STARFIRE design⁽¹⁾ developed at Argonne National Laboratory uses a static blanket concept, and tritium removal from this design concept has been analyzed in some detail^(2,3).

The third design concept, which employs moving solids, includes both conventional packed beds and fluidized beds. Tritium removal from the former has problems similar to those in the static beds, but the latter appears to have a number of advantages, not the least of which includes potential for recycling the bed material if effects due to radiation damage or tritium accumulation are unacceptable. Examination of the fluidized bed design was therefore included in the workscope of the current contract.

Mechanical design of two concepts for fluidizing the bed and carrying off excess heat and tritium is described in Section 9.1. One concept utilizes a high pressure stream of helium to remove the heat generated in the particles by neutron absorption; tritium did not diffuse fast enough into the gas stream so it was removed by moving the particles with the gas stream into an external collector. The other concept uses a low pressure helium stream to fluidize

the bed but removes the heat and the tritium by blowing the particles out of the blanket and into an external collector. The low pressure design was significantly simpler to construct and promised improved tritium breeding so it was chosen as the reference design for neutronic analyses presented in Section 9.2. In Section 9.3 the generation and diffusion of tritium in the Li_2O particles is analyzed; the analysis demonstrates that the low temperature existing in the blanket reduces the diffusion rate of tritium out of the particles below that necessary to prevent tritium buildup. Particle removal is therefore necessary to remove the tritium.

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9.1 MECHANICAL DESIGN

A solid lithium compound cooled by helium in a fluidized bed was the blanket concept selected for further study. The concept appeared to have the potential to produce a plausible design which would make a novel contribution to ICF reactor technology. Further, it was believed that in a porous but rigid solid lithium blanket, such as was proposed in the STARFIRE⁽¹⁾ reactor design, the grain boundaries would coalesce and the ceramic structure would crumble and sinter into a non-porous mass under the pulsed neutron radiation and high temperature. This condition would impede the transport of tritium gas from the solid causing the tritium inventory to increase too rapidly.

A fluidized bed is formed by forcing a fluidizing gas through thousands of small holes in the bottom of a containment vessel filled with a powder. The powder particles, when agitated by the passage of the gas, behave much like a liquid. They maintain a level surface, flow downhill, and squirt out through a hole located below the surface.

The initial work on the design of a fluidized bed blanket showed that two fundamentally different approaches could be used. In the high pressure approach, helium gas at 800 psi not only fluidizes the particles but it also is used as the sole method of heat removal. In the low pressure approach helium gas at 50 psi fluidizes the particles but carries away only 1% of the absorbed neutron energy. The remainder of the energy is carried out of the blanket by bleeding off the hot particles to an auxiliary fluidized bed heat exchanger. Each of these approaches was developed into a conceptual design so that their advantages and disadvantages could be compared and a preferred approach could be selected.

9.1.1 Description of the High Pressure Fluidized Bed Blanket Design

The layout of the reactor with the high pressure fluidized bed blanket is shown in Figure 9-1. The reactor is a right circular cylinder sitting on end. The change from the spherical shape (which was selected for the original liquid lithium cooled dry wall reactor⁽²⁾) was made because fluidized beds

**Figure 9-1. Elevation View of the High Pressure Fluidized Bed Blanket
Showing Overlapping Fluidized Beds**

are gravity sensitive. They will only work in an upright position because the gas bubbles rise vertically through the fluid. The pellet triggering beams come in horizontally at the vertical center plane of the cylinder just as they did in the original spherical design.

Helium was selected for the fluid medium because of its chemical inertness, its freedom from neutron activation, and its relatively high specific heat compared to other gases. Unfortunately, it is necessary to raise the pressure of helium to quite high levels in order to obtain a density which will transport the heat without the reactor becoming unreasonably large. A pressure of 800 psi was selected in order to obtain a gas density suitable for heat transfer. This high pressure had a very large effect on the general layout of the reactor because the pressure had to be contained without placing more than two centimeters of wall thickness between the tritium breeding compound and the neutron source. The two centimeters was established as a design criteria in the neutronic studies which were conducted on the liquid lithium dry wall ICF reactor design.⁽²⁾

In the blanket design for this reactor the neutrons are absorbed directly in the fluidized bed formed by the particles of the solid lithium compound floating in the helium gas stream. The entire solid angle surrounding the reaction chamber therefore is surrounded with fluidized beds. For the blanket power density corresponding to a first wall inside radius of 10 meters, each fluidized bed must be approximately two meters deep. In this depth, the fluidizing coolant gas acquires the desired temperature rise from 300°C to 475°C. These temperatures were selected in the STARFIRE⁽¹⁾ design as being about optimum. The 475°C was set by the need to retain adequate strength in the structural material while the 300°C was set by the desire for an efficient steam cycle for power conversion.

The application of a fluidized bed to a neutron absorbing blanket requires the fluidized bed to operate in a manner which is quite different from most fluidized bed applications. In the blanket, the heat enters the bed largely at one side because the absorption of neutron energy drops off exponentially in the radial direction through the bed. The tendency of a fluidized bed to

operate at a uniform temperature throughout distributes the energy absorbed at one side and thereby avoids hot spots where the temperature is above the safe operating temperature for the steel containers. This function is accomplished in solid bed designs, such as STARFIRE, by using an increased density of water cooled tubes near the first wall surface. This is a much more complex mechanical construction than a fluidized bed.

It is inherent in fluidized bed devices that considerable free space exist above the surface of the fluidized bed. If this is not done, there will be excessive carryover of solid particles in the exiting helium. Figure 9-1 shows how individual fluidized beds are stacked one above the other in two concentric annuluses to eliminate the neutron streaming paths through the free space above the beds. The height of the bed varies with the energy density at each location to insure that the helium leaves the bed at 475°C.

The top view of the cylindrical reactor in Figure 9-1 shows that each annulus is divided radially into 136 separate compartments. This division is made necessary by the high pressure of the helium and the requirement that there be no more than two centimeters of steel between the pellet and the breeding material. The rounded noses of the compartments at the inside of the blanket are one centimeter thick and are subjected to a hoop stress of 20000 psi. The flat compartment sides at the outer surface of the blanket have an outward force against the hoop tension bands. The sidewalls of the compartments can be looked on as tension membranes which tie the inner cylindrical noses and the outer compartment side together. The net outward force which is developed is supported by a cylindrical hoop tension band which wraps around the entire reactor.

The compartment shape shown in the top view of Figure 9-1 exists at all horizontal cross sections. The compartments are really tall tanks which extend from the bottom to the top of the reactor. The interior dividers which form the stacked individual fluidized beds shown in Figure 9-1 are all contained inside the pressurized tank and thus need only be designed to

support the weight of the beds and the pressure drop across a bed. This pressure will run from 3 to 10 psi, a negligible pressure compared to the 800 psi mean pressure in the tank.

The use of individual tanks is essential for maintenance and repair. While the blanket is designed to last the life of the plant, provision must always be made for an unplanned failure which requires the replacement of a major component. In this design an individual tank with its contents weighs only 100 tons so that it can be transported by an overhead crane. All connections to the individual fluidized beds are made at the outer periphery of the blanket where they can be disconnected remotely.

The 300°C coolant gas is brought to the individual fluidized beds through ten circumferential manifolds, one manifold at each annular level. Likewise, the 475°C exit gas is collected in ten circumferential exhaust manifolds. The gas velocity is maintained high enough in these exhaust manifolds to guarantee that the fines which escape from the beds are transported to central cyclone and electrostatic separators which remove the solids before the gas goes to the steam generator. These gas manifolds are located at elevations which are in between the elevations of the hoop tension rings so that connections from the manifolds to the beds do not need to penetrate the rings. There are no connections between the tanks inside the hoop tension band.

Altogether there will be 1360 individual fluidized beds. Each bed will require an inlet and an outlet for the fluidizing gas. It will also require one small diameter pipe for the introduction of new powder and one small diameter pipe for the drainage of spent powder. Also required will be one pressure tap near the surface of the bed to give information on whether additional powder should be added. The maintenance of the proper bed height and gas flow to each of the individual beds would be computer controlled to provide proper fluidization and a constant temperature output.

The top view of the reactor in Figure 9-1 shows the horizontal tank fluidized beds which form the top and bottom of the cylinder. The top and bottom areas must be divided into compartments which are only 20 inches wide so that the

one centimeter thick walls on the half cylinders which form the tops and bottoms of the compartments will not be stressed over 20000 psi by the 800 psi helium. The vertical cross section through the top of the cylinder in Figure 9-1 shows the shape of the compartments. Again, because the compartments are not round, they must be surrounded by a tension band to prevent them from expanding when pressurized. The fluidized beds in this case are long and thin and are of variable length, but all are of the same thickness. The beds are two meters deep to assure that practically all of the neutrons are absorbed.

A water cooled first wall to replace the liquid lithium first wall of the original dry wall design is shown in Figure 9-1. Vertical water tubes would run from the horizontal center plane to the top of the cylinder and from the horizontal center plane to the bottom of the cylinder. The water would be at 300°C and 15.2 MPa (2000 psi) as is standard practice for pressurized water fission power plants. The tubes would be 3.5 cm mean diameter with a wall thickness of .18 cm. These tubes would be electron beam welded to a .6 cm thick support wall which would also be the vacuum boundary. The effective steel thickness of this first wall is just about the additional 1 cm of the allowable 2 cm which is fixed by the neutronics requirements. The remaining one centimeter is given to the fluidized bed containment wall. Additional structure will be required to prevent this first wall from buckling under atmospheric pressure. The first wall will consist of an upper and a lower half cylinder completely independent of the blanket structure so that it can be removed for maintenance exactly as described in the original lithium cooled reactor design.

In contrast to the lithium cooled design where the first wall and the blanket operated at the same temperature, the fluidized bed design operates with the water cooled first wall at 300°C while the fluidized bed is at 475°C. A layer of thermal insulation will be placed between the two to prevent additional heat input to the first wall which is already hard pressed to absorb the surface heat flux.

In the operation of this high pressure design, the particles remain permanently in the bed. However the particle size is being continuously reduced by the erosion in the bed. The fines that are produced are continuously blown out with the fluidizing gas and are separated from the gas in cyclone separators. These fines are further pulverized to the .1 to 1 μ m grain size and are heated to speed the removal of the tritium by diffusion before the elemental grains are sintered into new particles of the size required for the proper operation of the fluidized bed.

9.1.2 Description of the Low Pressure Fluidized Bed Blanket Design

The low pressure fluidized bed blanket is shown in the elevation view of Figure 9-2. As in the high pressure design the reaction chamber is a right circular cylinder standing on end. The blanket which forms the cylindrical wall of the chamber is made up of thirty-six tall steel tanks which run the full thirty-two meter height of the chamber. Each of these tanks forms the containment for an individual fluidized bed of sintered lithium oxide pellets (which have a maximum diameter of one millimeter) suspended in helium gas at 50 psi. The blanket has an average radial thickness of two meters. Neutrons give up their energy to the suspended particles which are in turn continuously withdrawn at 475°C from the top of each tank to carry the heat to the fluidized bed heat exchangers shown in Figure 9-3 which serve as steam generators. The cooled particles at 300°C are then returned to the bottom of the fluidized bed tank by a screw feeder. About 1% of the neutron energy is carried away by the fluidizing gas to a separate heat exchanger. Horizontal fluidized beds two meters deep, contained in tanks two meters wide, form the top and bottom of the cylindrical reaction chamber. Again the heat is removed from the fluidized beds by withdrawing the hot particles. The 300°C particles are fed into one end of the horizontal fluidized bed and flow by gravity like a liquid to be spilled out at the far end at 475°C. The particles are transported by fluidized bed piping to the fluidized bed heat exchangers. A small portion of the circulating particle stream will be withdrawn continuously for heating to a high temperature to drive off the tritium. These particles will be pulverized, mixed with the fines separated from the

Figure 9-2. Elevation View of the Low Pressure Fluidized Bed Blanket

Figure 9-3. Fluidized Cooler for Hot Alumina Particles

fluidizing gas and resintered into particles of the size required for proper operation of the fluidized beds. The first wall for the low pressure blanket will be essentially as already described for the high pressure blanket.

9.1.3 Comparison of the Low and High Pressure Designs

Many of the advantages of fluidized beds over static beds for a neutron absorbing blanket are common to both designs, as indicated below:

- A. The particles of the lithium compound which absorb the neutrons and become hot are always in intimate contact with the helium coolant. In contrast to the static bed design, the fluidized bed does away with the hundreds of thousands of tiny tubes which would have to be placed in solid but porous blocks of lithium compound in order to provide a low thermal drop between the solid and the coolant. Also, the very complex manifolding required to bring the coolant to all these small coolant tubes is eliminated. The thousands of welded tubes needed in the static bed design give a very high probability for leaks. In the tank design for the fluidized beds, there are no delicate welds and the number of welds is small. Leaks if they occur would be outside the plasma vacuum space.
- B. A fluidized bed is characteristically isothermal because the agitated particles cause the bed to have a very high thermal conductivity. Thus, even though the neutron absorption is highest on the tank side facing the exploding pellet, the temperature is nearly uniform throughout the bed. This permits a higher average blanket temperature for a given inner tank wall temperature.
- C. The particles of the lithium compound are bled off continuously to permit the removal of the tritium. This greatly reduces the tritium inventory.

D. Fluidized beds are a commonly used technology in the chemical processing industry. They have formed the basis for the catalytic cracking of gasoline since the early 1940s. Fluidized bed design practices are well established and the cost of developing a fluidized blanket will be low.

The advantage of the high pressure design is that auxiliary fluidized bed heat exchangers are not required. The heat is removed from the fluidizing gas in a relatively standard boiler which is much simpler than a fluidized bed heat exchanger.

The advantage of the low pressure design is its simplicity. The fluidized bed containment tanks are large and few in number. Each vertical tank is a single fluidized bed compartment while the high pressure design requires that the same size tank be fitted with dividers to create small independent beds. Since the low pressure tank walls are thinner, they will absorb fewer neutrons, thus giving a larger tritium breeding ratio. Only 56 independent fluidized beds are required instead of the 1360 required for the high pressure design. This greatly simplifies the manifolding and reduces the cost of the controls for the bed depth and gas flow.

Since the advantages of the low pressure design appear to outweigh those of the high pressure design, the low pressure design was selected as the basis for the neutronic analysis. Lithium oxide was selected for the low pressure blanket for neutronics reasons while the design of the high pressure blanket was based on lithium aluminate as used in the STARFIRE blanket. The change in the fluidized solid has very little effect on the design of the fluidized beds since the density and specific heat of the two solid compounds are not much different, as seen from the comparison below:

	Specific Heat	Specific Gravity
Lithium Aluminate	1.6 J/g-K	2.04 g/cm ³
Lithium Oxide	2.6 J/g-K	2.01 g/cm ³

Since there is no information available on the fluidization behavior of the two compounds, the sphericity factor was assumed to be 0.6 for both.

The calculations which support the design of the fluidized beds used in the blankets are given in the following section.

9.1.4 Calculations Supporting the High Pressure Design

MINIMUM MASS FLOW FOR FLUIDIZATION

The fluidizing gas is forced through thousands of small holes in the bottom of the containment for the powder in order to cause the powder to fluidize. The mass velocity for minimum fluidization, G_{mf} , is the most basic design relationship; it is given in references 3 and 4 and reproduced here.

$$G_{mf} = \frac{688 D_p^{1.82} [\rho_f (\rho_s - \rho_f)]^{0.94}}{\mu^{0.88}} \quad (1)$$

G_{mf} is in pounds per hour per square foot

D_p is the particle diameter in inches

ρ_f and ρ_s are the gas and particle densities in pounds per cubic foot

μ is the viscosity of the gas in centipoise.

For the reference design, the particle diameter, D_p , was assumed to be .010 inch (.254 mm or 254 microns). This diameter is within the range of the grain size, (.1 to 1.0 mm) selected for the STARFIRE breeder material and is a commonly used size for fluidized beds.

The density of the particle is taken as 127 lb/ft³. This was obtained from the information on the LiAlO₂ particles given in the STARFIRE reports. The

solid material has a density of 3.4 g/cm^3 . In the processing to provide porosity for tritium escape, the grains are sintered into a block which only 60% of this density or 2.04 g/cm^3 . This is the same as 127 lb/ft^3 .

The viscosity of helium is independent of temperature and pressure and is given in handbooks as .03174 centipoises.

The density of helium, ρ , is calculated from the characteristic equation for a perfect gas, $\rho V = wRT$. The temperature used for this calculation was taken as the mean of the selected helium inlet and outlet temperatures.

$$T = \frac{300 + 475}{2} = 387.56 = 729.5 \text{ F}$$

$$T = 729.5 + 460 = 1189.5 \text{ Rankine}$$

$$p = 800 \times 144 = 115,200 \text{ lb/ft}^3$$

$$R = 386 \text{ in English units}$$

$$\rho_F = \frac{W}{V} = \frac{p}{RT} = \frac{115,200}{386 \times 1190} = 0.250 \text{ lb/ft}^3$$

Substituting in equation (1) gives:

$$G_{mf} = \frac{688 \times 0.01^{1.82} [0.250 (127 - 0.250)]^{0.94}}{.0317^{0.88}}$$

$$= 84.5 \text{ lb/hr} \cdot \text{ft}^2$$

Fluidized beds normally operate with gas flows between two and fifteen times this minimum value.

BED HEIGHTS FOR EACH ANNULUS

The radial thickness of the inner and outer annular fluidized beds was arbitrarily selected at three meters for a trial design. Since this thickness produced a reasonable design, it has been retained. Certainly a lot of work could be done to determine a more optimum thickness, but in view of absence of the fluidization characteristics of the lithium compound, a more refined design is not justified.

For the trial design, a mass flow rate of 8.5 times G_{mf} was selected since this is half-way between the 2 and 15 times G_{mf} which are the edges of the range represented in common practice.

The inner radius of the inside annulus was selected as eleven meters since the first wall is located at 10.0 meters and some space is required between the first wall and the blanket for the manifolds for the first wall coolant.

Neutronics calculations show that of the 3500 MW_{th} to be developed by the reactor, 68% will go into the blanket. Also, there will be an energy multiplication in the blanket of about 1.2. The total power to be absorbed by the blanket is then:

$$Q = 3500 \times .68 \times 1.2 = 2856 \text{ MW}$$

Since the pellet will radiate equally in all directions, the power density leaving the surface of a sphere 11.0 meters in radius is:

$$\frac{2850}{4\pi r^2} = \frac{2850}{4 \times \pi \times 11^2} = 1.88 \text{ MW/m}^2$$

The procedure for calculating the height of a bed will be illustrated by the calculation for the outer annulus just above the horizontal plane at the vertical center of the reactor (see Figure 9-1).

The area of the floor of this annulus which has an inside diameter of 14 m and outside diameter of 17 m is:

$$\begin{aligned} A_B &= \pi (17^2 - 14^2) \\ &= 292 \text{ m}^2 \\ &= 3140 \text{ ft}^2 \end{aligned}$$

This is the area of the bottom of the fluidized bed and the area of the perforated plate which distributes the helium to the bed. The mass flow rate for the annulus is then:

$$\begin{aligned} G_f &= G_{mf} \times 8.5 \times 3140 \\ &= 84.5 \times 8.5 \times 3140 \\ &= 2,260,000 \text{ lb/hr} \\ &= \frac{2,260,000}{3600 \times 2.2} = 285 \text{ Kg/s} \end{aligned}$$

The heat rate, Q, which can be absorbed by this flow rate is:

$$Q = \text{Kg/sec} \times c_p \times \Delta T$$

where c_p is the specific heat for helium

$$\begin{aligned} c_p &= 1.25 \text{ BTU/lb/}^\circ\text{F} \\ &= 1.25 \times 1055 \times 2.2 \times 9 \end{aligned}$$

$$c_p = 5222 \text{ joules/kg} \cdot ^\circ\text{K}$$

The inlet and outlet temperatures for the helium coolant were established previously as 300°C and 475°C respectively. Therefore:

$$\Delta T = 475 - 300 = 175^{\circ}\text{C}$$

$$Q = 285 \times 5200 \times 175$$

$$= 259 \text{ MW}$$

The surface area of the hypothetical 11 meter radius sphere (see Figure 9-1) through which this amount of power flows is:

$$\frac{Q}{\text{Surface Power Density}} = \frac{259}{1.88} = 137 \text{ m}^2$$

The formula for the area of a spherical segment where the horizontal plane at the vertical center is the base is:

$$A_s = 2\pi rh$$

where r is the radius of the sphere

h is the height above the base plane of the intersecting parallel plane.

$$h = \frac{A_s}{2\pi r}$$

$$= \frac{137}{6.28 \times 11} = 1.98 \text{ m}$$

The angle with respect to the base plane, ψ , at the circle of intersection is:

$$\psi = \sin^{-1} \frac{h}{r} = \sin^{-1} \frac{1.98}{11} = 10.37 \text{ degrees.}$$

All of the radiant energy which is absorbed in the annulus under consideration is emitted from the sphere in this angle.

The height of the bed at the inner radius of the annulus of 14 m is:

$$\begin{aligned}h_B &= 14 \times \tan \psi \\ &= 2.56 \text{ meters.}\end{aligned}$$

This is a reasonable height or depth for a fluidized bed. All of the bed heights shown in Figure 9-1 were determined in a similar manner. It should be noted in Figure 9-1 that the neutrons radiating from the center of the sphere must pass through a full three meters of bed thickness in either the inner or the outer annulus.

TANK NOSE RADIUS

High pressure fluids are contained with the least thickness of material at a given stress in a round pipe. The noses for the vertical tanks should therefore be round in order to reduce the thickness to a minimum to give good neutronic performance. A thickness of one centimeter has been proposed for the tanks from neutronics considerations and 20,000 psi is a reasonable material stress at the operating temperature. The radius of the tank nose to meet these specifications is calculated as follows.

The equation for hoop stress in a cylinder is:

$$s = \frac{p r}{t}$$

Where p is the pressure in psi = 800

r is the mean radius of the cylinder

t is the thickness of the cylinder = 1 cm = .40 in

s is the allowable stress in psi 20,000

$$r = \frac{st}{p}$$

$$r = \frac{20000 \times .4}{800} = 10 \text{ inches} = .254 \text{ meters.}$$

This nose radius is shown for all of the tanks in Figure 9-1. It determines the total number of tanks required as follows:

$$\text{Number of tanks} = \frac{\text{Circumference of inner wall of inner annulus}}{\text{Diameter of tank nose}}$$

$$136 = \frac{2\pi \ 11.0 \times 39.4}{20}$$

The sidewalls of the tanks will have the same stress because the inside wall is to be suspended from the outside wall assuming that the convoluted inside wall is not capable of supporting any compressive load.

HOOP TENSION BAND

The equation for the hoop tension in the outer wall of a pressurized annulus with the inner wall suspended on the outer wall is:

$$P = q_o (r_o - r_i)$$

Where q_o is the unit force per inch of circumference per inch of axial length on the outer cylinder (q_o equals the fluid pressure)

r_o is the radius of the outer cylinder

r_i is the radius of the suspended inner cylinder.

For the reactor blanket design of Figure 9-1

$$q_o = 800 \text{ psi}$$

$$r_o = 17 \text{ m} = 670 \text{ in}$$

$$r_i = 11 \text{ m} = 433 \text{ in}$$

$$P = 800 (670 - 433)$$

$$P = 189,600 \text{ lb}$$

For a tensile stress of 20,000 psi, the hoop tension band will have to have a thickness of:

$$t = \frac{P}{s} = \frac{189,600}{20,000} = 9.46 \text{ in}$$

It is interesting to note that this thickness would have to be 26.8 inches if the interior wall were not suspended from the outer wall.

PRESSURE DROP TO FLUIDIZE THE BED

The equation for the pressure drop across a bed to cause fluidization is:

$$\Delta p = L (1 - \epsilon) (\rho_s - \rho_F)$$

Where Δp is the pressure in lb/ft^2

L is the bed height in ft

ϵ is the void fraction

ρ_s is the particle density in lb/ft^3

ρ_F is the fluid density in lb/ft^3

Use $L = 2.56 \text{ m (8.40 ft)}$, $\rho_s = 127 \text{ lb/ft}^3$ and $\rho_F = 0.250 \text{ lb/ft}^3$ from the calculation of the height of the first annulus above the horizontal plane. The value of ϵ will be somewhere near 0.4. Its exact value can be established only by testing the characteristics of the powder.

$$\begin{aligned} \Delta p &= 8.40 (1 - .4) (127. - 0.25) \\ &= 640 \text{ lb/ft}^2 \\ &= 4.44 \text{ psi} \end{aligned}$$

The pressure drop across the bed is thus small compared to the 800 psi required to increase the density of the helium.

PIPE DIAMETER FOR HELIUM INLET AND OUTLET

In the sample calculation of the bed height for an annulus, the mass flow rate to the annulus was calculated at 2,260,000 lb/hr. The flow rate for each of the 136 individual fluidized beds in the outer annuli is then

$$\frac{2,260,000}{136} = 16,600 \text{ lb/hr} = 4.61 \text{ lb/sec.}$$

From the same sample calculation, the mean gas density was calculated at .25 lb/ft³. The volume flow is thus:

$$\frac{4.61}{.25} = 18.4 \text{ ft}^3/\text{s}$$

Assume that the helium velocity is to be held to mach .07 to keep the pumping power low. The helium velocity at mach 1 is calculated by the equation

$$a = \sqrt{k g R T}$$

a = mach 1 velocity in ft/sec

k = ratio of specific heats

g = acceleration of gravity = 32.2 ft/s²

R = gas constant = 386

T = mean gas temperature, Rankine = 1190

$$a = \sqrt{1.659 \times 32.2 \times 386 \times 1190}$$
$$= 5007 \text{ ft/sec}$$

Allowable velocity = .07 x 5007 = 350 ft/sec.

$$\text{Pipe cross sectional area} = \frac{18.4}{350} = .0526 \text{ ft}^2 = 7.57 \text{ in}^2$$

$$\text{Pipe inside diameter} = \frac{\sqrt{4 \times 7.57}}{\pi} = 3.10 \text{ in}$$

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9.2 Neutronic Analysis Of Improved Fluidized Bed Design

One dimensional spherical geometry coupled neutron-photon transport calculations were performed for the new fluidized bed blanket design. The primary objective of these calculations was to provide blanket dimensional requirements for adequate tritium breeding and neutron energy deposition. A tritium breeding ratio goal of about 1.2 was established to account for calculational uncertainties, chamber penetrations, and tritium losses. Total energy deposition in the energy conversion part of the first wall and blanket zones should be much less than the recirculating power fraction of the ICF plant. In the absence of this information, an energy loss rate goal less than 1% was rather arbitrarily set.

An additional calculated parameter is the radiation damage lifetime of the first wall. It is important to demonstrate that the reactor cavity is sized to provide a first wall lifetime of at least five years. This insures a first wall replacement schedule consistent with the utility maintenance cycle for major plant shutdown and overhaul.

9.2.1 Calculational Model

The calculations were performed with the codes and cross section data base described earlier⁽¹⁾. The burning pellet was approximated by an equimolar mixture of deuterium and tritium with a density-radius product of 3g/cm^2 . Neutrons were started uniformly and isotropically inside this source. This source provides a neutron energy fraction of approximately 68%, with the remaining 32% appearing as a surface heat load to the first wall.

The one-dimensional spherical geometry representations of the side, top, and bottom of the cylindrical reaction chamber are listed in Tables 9-1 through 9-3. It was necessary to separate the solution into three parts because of the different configuration of the cylinder components. Local results

TABLE 9-1
NEUTRONICS CONFIGURATION OF CHAMBER SIDE STRUCTURE
(Spherical Geometry)

<u>Zone</u>	<u>Composition</u>	<u>Outer Radius (cm)</u>
1	Pellet Source	0.02
2	Chamber (Vacuum)	1000.0
3	First Wall Structure	1000.7
4	First Wall Support	1001.3
5	Plenum Zone	1050.0
6	Fluidized Bed Wall	1051.0
7	Fluidized Bed	1351.0
8	Fluidized Bed Wall	1355.0
9	Graphite Reflector	1375.0
10	Bulk Shield	1675.0

TABLE 9-2
NEUTRONICS CONFIGURATION OF CHAMBER TOP STRUCTURE
(Spherical Geometry)

<u>Zone</u>	<u>Composition</u>	<u>Outer Radius (cm)</u>
1	Pellet Source	0.02
2	Chamber (Vacuum)	1000.0
3	First Wall Structure	1000.7
4	First Wall Support	101.3
5	Plenum Zone	1050.0
6	Front Nose of Beds	1150.0
7	Distribution Plate	1150.5
8	Fluidized Bed	1350.5
9	Sidewall Bed Structure	1550.5
10	Back Nose of Beds	1650.5
11	Graphite Reflector	1670.5
12	Bulk Shield	1970.5

TABLE 9-3
NEUTRONICS CONFIGURATION OF CHAMBER BOTTOM STRUCTURE
(Spherical Geometry)

<u>Zone</u>	<u>Composition</u>	<u>Outer Radius (cm)</u>
1	Pellet Source	0.02
2	Chamber (Vacuum)	1000.2
3	First Wall Structure	1000.7
4	First Wall Support	1001.3
5	First Wall Plenum	1050.0
6	Front Nose of Beds	1150.0
7	Sidewall Bed Structure	1350.0
8	Fluidized Bed	1550.0
9	Distribution Plate	1550.5
10	Back Nose of Beds	1650.5
11	Graphite Reflector	1670.5
12	Bulk Shield	1970.5

(volumetric heating, radiation damage) are provided by the separate solutions. Global results (tritium breeding, energy production) are solid angle weighted averages.

9.2.2 Composition

FIRST WALL

The ICF fluidized bed design has a separately cooled and supported first wall structure. While the use of two different coolants complicates the plant energy conversion system, it is unavoidable. The fluidized medium (Li_2O) particles cannot be used to remove the high first wall surface heat load of X-rays and pellet debris ions without generating high temperatures and thermal stresses. The average surface heat load is 0.6 MW/m^2 , with a peak value of 0.9 MW/m^2 (at the surfaces closest to the pellet).

The first wall consists of pressurized water cooled corrugated panels, and is an adaptation of the STARFIRE⁽²⁾ design. The average surface heat load is 0.7 MW/m^2 for STARFIRE, close to the present value of 0.6 MW/m^2 . The dimensions of the STARFIRE first wall were therefore retained in the neutronic calculations. In particular, the corrugated surface, which faces the pellet, consists of a 1.5mm HT-9 substrate coated with a 1.0mm layer of tantalum.⁽³⁾ The 0.7mm thick first wall zone (zone 3 in Tables 9-1,2,3) then consists of 32 v/o water, 21 v/o HT-9, 14 v/o tantalum and 23 v/o void. A plenum zone approximately 50 cm wide is included behind the first wall structure to contain all of the piping and manifolding required for the pressurized water coolant distribution system.

FLUIDIZED BED

Lithium oxide (Li_2O) at 70% of theoretical density and 50% packing fraction was selected for the fluidizing bed material because of the difficulty in achieving adequate tritium breeding in a low density fluidized bed blanket, identified earlier⁽¹⁾. The use of lithium aluminate (LiAlO_2), an optional blanket material, requires the addition of a beryllium multiplier, as well as

enrichment of the lithium in ^6Li . The former introduces a number of issues, including added design complexity, safety, and resource availability. The latter has a significant cost impact.

The side walls and front nose structure of the fluidizing bed tanks were assumed to be 0.635 cm thick HT-9, with the noses hemispherical in shape and having a diameter of 2 meters. No other structure was included in the fluidized bed zones.

REFLECTOR AND SHIELD

A 20 cm thick graphite reflector with 5 v/o HT-9 structure was included in the calculations. Void space was included for coolant, but no coolant was specified. The choice of coolant will depend upon a decision regarding energy recovery in the reflector zone. The reflector was followed by a three meter thick ordinary concrete shield. This was shown in earlier studies to provide an adequate bulk biological shield for the reference chamber size and D-T power output.

9.2.3 Results

TRITIUM BREEDING

The tritium breeding ratios (TBR) and neutron absorption distributions of the reaction chamber components are listed in Table 9-4. The solid angle weighted global TBR is 1.27, exceeding the design goal of 1.2 by a substantial margin. One reason for this improved breeding performance, compared to the earlier fluidized bed design, is the lower structural volume fraction. This results in a parasitic absorption rate of less than 10%.

Figure 9-4 shows the cumulative tritium breeding ratio through the side wall fluidized bed. Similar curves apply to the top and bottom beds. It can be seen that the TBR rises rapidly at first, slowing down above about 80 cm in bed thickness, and then approaching the full bed thickness (300 cm) value of 1.28 in a near asymptotic fashion. The target value of 1.20 occurs at a thickness of approximately 120 cm. Thus, on the basis of TBR requirements, it

TABLE 9-4
TRITIUM BREEDING RATIOS AND NEUTRON ABSORPTION DISTRIBUTIONS

	<u>Neutron Absorptions (Percent)</u>				<u>Li₂O</u>
	<u>TBR</u>	<u>HT-9</u>	<u>Ta</u>	<u>Water Coolant</u>	
Side Wall	1.28	4.1	6.2	0.1	89.6
Top Wall	1.26	5.7	5.6	0.1	88.4
Bottom Wall	1.25	6.7	4.6	0.1	88.6

TABLE 9-5
ENERGY DEPOSITION DISTRIBUTIONS

	<u>Energy Deposited (MW)</u>			<u>Total</u>
	<u>First Wall Structure</u>	<u>Fluidized Bed</u>	<u>Reflector & Shield</u>	
Side Wall	127	1783	0.38	1911
Top Wall	40	596	2.8	639
Bottom Wall	37	599	1.3	637
Totals	204*	2978	4.5	3187
Total Thermal Energy: 4307 MW				

*1120 MW of X-ray and ion energy must be added to this to determine first wall heat removal system requirements.

appears that a considerable reduction in fluidized bed thickness is possible. However, an additional neutronics requirement is adequate energy deposition in the blanket region, and this is addressed below.

ENERGY DEPOSITION

The energy deposition distribution in the various chamber components is listed in Table 9-5. It can be seen that only 4.5 MW, or about 0.1% of the total available energy is deposited outside of the first wall and blanket. This is much lower than the goal of 1% and suggests, in a fashion analogous to that for tritium breeding, that the blanket thickness can be reduced. Figure 9-2 is a plot of the total available neutron energy deposited in the blanket as a function of blanket thickness. It is similar in shape to the TBR distribution of Figure 9-4. This is not surprising since 65% of the total deposited neutron energy is due to tritium producing reactions with ${}^6\text{Li}$ and ${}^7\text{Li}$.

It can be seen in Figure 9-5, that at a blanket thickness of 120 cm, which is adequate for tritium breeding, nearly 97% of the available neutron energy is deposited in the first wall and blanket. If the surface heat deposition of 1120 MW is included, 98% of the total energy is deposited. Moving the graphite reflector adjacent to this thinner blanket would raise this close to the 99% goal.

RADIATION DAMAGE LIFETIME

The average first wall neutron loading is 1.3 MW/m^2 , rising to a maximum value of 1.9 MW/m^2 at the surfaces nearest to the chamber center. Fluence lifetime under ICF reactor conditions for HT-9 is not well defined, but we can probably assume it to be close to that of titanium modified 316-SS (prime candidate alloy). Under low thermal stress steady state conditions this has been estimated to be 20 MWy/m^2 . These conditions are reasonably well satisfied in the present case. The wall thickness has been chosen to be that identical to the thin walled STARFIRE design, and temperature fluctuations between pellet microexplosions has been estimated to be less than 50°C . On this basis the expected first wall lifetime is therefore approximately 10 years.

Figure 9-4. Tritium Breeding Ratio Vs. Fluidized Bed Thickness

Figure 9-5. Neutron Energy Deposition Fraction Vs. Fluidized Bed Thickness

Another basis for estimating radiation damage lifetime is to examine displacement damage and helium production rates. These were calculated to be 10.7 DPA/year and 104 appm He/year, respectively, at 70% plant availability. Assuming again that we may use results for 20% cold worked 316 SS, limits of 165 DPA and 500 appm He at an operating temperature of 500°C have been suggested.⁽⁴⁾ This results in a first wall lifetime of 15 years due to displacement damage and 5 years due to helium production. However, the ENDF/B-IV helium production cross sections used in the calculations are probably high. Calculations by Meier⁽⁵⁾ for the HYLIFE concept gave helium production rates a factor of two lower when the ENDL library was used.

From the above considerations we may conclude that, based on available data and calculations, the first wall radiation lifetime damage goal of five years is satisfied for the present ICF fluidized bed dry wall design.

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9.3 Generation and Diffusion of Tritium in Lithium Compounds

This section describes the analysis of tritium generation and removal from solid lithium compounds in the fluidized bed design. Tritium removal from the STARFIRE⁽¹⁾ blanket was reviewed since four of the five steps in the tritium recovery process considered in the ANL analysis will also occur in the Westinghouse fluidized bed design. The results and conclusions of the STARFIRE study, however, do not apply directly to the Westinghouse concept, since the latter sees a pulsed neutron flux and operates at a lower temperature than the former. The analytical models developed to evaluate tritium inventory and release in the STARFIRE blanket are also not appropriate for an analysis of the Westinghouse design, primarily because of the assumption of continuous tritium generation in the STARFIRE design. The model for calculating diffusive inventory therefore had to be modified to show the time dependence; the revision is described below in Section 9.3.1.

The blanket design proposed for the STARFIRE tokamak reactor is shown schematically in Figure 9-6. Pellets of α -LiAlO₂, with grain radii ranging between 0.1 and 10 μ m and a fine porosity, are packed in a bed that contains a regular hexagonal array of coolant tubes. Low pressure ($\sim$ 0.05 MPa) helium gas is circulated through 2mm diameter channels formed in the breeder material.

Tritium release in the STARFIRE blanket involves five steps:

- A. Bulk diffusion in the grain,
- B. Desorption of T₂O at the grain surface,
- C. Migration through interconnected grain boundary porosity to the particle surface,
- D. "Percolation" through the porosity in the packed bed of particles, and

Figure 9-6. Schematic Diagram of STARFIRE Blanket Concept (Taken from Reference [1])

E. Convective mass transfer out of the blanket in the helium processing stream.

Steps A through C and step E will occur in the fluidized bed. All, however, proceed under operating conditions which are sufficiently different from those of the STARFIRE blanket that a re-analysis is necessary. The impact of the bulk diffusion step on tritium release and inventory changes when tritium generation is pulsed rather than continuous, since, under certain conditions, a steady-state diffusive inventory may not be established. Desorption of T_2O at the grain surface may not dominate at the lower operating temperature, as it does at high temperature. The primary species detected in low temperature reactor experiments is HTO rather than T_2O .⁽²⁾ Tritium release from Li_2O single crystals during irradiation at low temperatures occurs by a recoil mechanism, rather than thermal diffusion.⁽³⁾ This has been observed for irradiations between 100 and 300 C. The maximum temperature for the fluidized bed is 475 C, lower than for the STARFIRE blanket, and it may be anticipated that the recoil mechanism affects release. For these reasons, the results and conclusions of the STARFIRE analysis do not apply directly to the Westinghouse design.

9.3.1 Time Dependent Bulk Diffusion Analysis

Evaluation of the diffusion coefficient, D, using two different empirical formulas leads to values which are different by a factor of 10^7 . The time dependent analytic model will be used to estimate D.

There are two contributions to the tritium inventory in the solid-breeder material: one that results from the thermodynamic solubility of tritium in the ceramic and one that arises because of the competition between tritium production and release processes. The latter depends on the rate-limiting release mechanism and reactor operating conditions. In the analysis of tritium release for the STARFIRE design, diffusion of tritium through the grain to the grain boundary was found to be the rate-limiting step. The contribution to the tritium inventory was calculated assuming that a balance

between tritium generation and diffusion to the grain boundaries had been attained and that, therefore, steady-state diffusion equations were appropriate for the analysis. This is a reasonable assumption for the STARFIRE design, but is not for an ICF design, where tritium production is pulsed, as shown schematically in Figure 9-7. Pulse duration times are of the order of 10 to 100 ns, and anticipated repetition rates are between 1 and 20 s^{-1} . The analysis of the diffusive inventory must be modified for these conditions.

We assume that grains are spherical with radius R and that the tritium generation rate, $\dot{I}(t)$, is $\dot{n}$ for $0 \leq t \leq \tau$, where τ is the pulse width, and that $\dot{I}(t)$ is 0 for $\tau < t \leq \delta$, where δ is the cycle time. During the first cycle for $t \leq \tau$, the tritium concentration $c(r,t)$ in the particle satisfies the differential equation

$$\frac{\partial c(r,t)}{\partial t} = D \frac{\partial^2 c(r,t)}{\partial r^2} + \frac{2}{r} \frac{\partial c(r,t)}{\partial r} + \dot{n} \quad [1]$$

where D is the diffusion coefficient, with the boundary conditions $c(r,0) = 0$ and $c(R,t) = 0$. For $t > \tau$, $c(r,t)$ satisfies the equation

$$\frac{\partial c(r,t)}{\partial t} = D \frac{\partial^2 c(r,t)}{\partial r^2} + \frac{2}{r} \frac{\partial c(r,t)}{\partial r}, \quad [2]$$

subject to the boundary condition $c(R,t) = 0$ and with an initial distribution $c(r,\tau)$ that is the solution to Eq. [1] evaluated at $t=\tau$. At the beginning of the second pulse, the tritium concentration again satisfies Eq. [1], but with an initial concentration profile $c(r,\delta)$ obtained from Eq. [2]. In this manner, Eqs. [1] and [2] can be solved sequentially with changing boundary conditions, and the concentration profile obtained as a function of time for various values of τ , δ , $\dot{n}$, and temperature.

Figure 9-7. Schematic of Tritium Production Rate Versus Time in ICF Reactor

Without explicitly solving for $c(r,t)$ by this procedure, two types of limiting behavior can be anticipated, depending on the value of the diffusion coefficient D . For a particle with radius $R \ll 2\sqrt{Dt}$, the tritium concentration will peak at the end of the pulse and decay during the off time, as shown in Fig. 9-8a. For $R \gg 2\sqrt{Dt}$, the tritium concentration will remain unchanged during the off time, and build when the pulse is on, as shown in Fig. 9-8b.

No experimental data is available on tritium diffusion in Li_2O and LiAlO_2 for conditions of interest. Smith et. al.⁽⁴⁾ used an estimated diffusivity given by

$$D = 3.26 \exp (-5.72 \times 10^4/RT) \text{ cm}^2/\text{s} \quad [3]$$

for the STARFIRE analysis. Kudo and Okuno⁽²⁾ have obtained a diffusion coefficient

$$D = 0.019 \exp (-2.48 \times 10^4/RT) \text{ cm}^2/\text{s}, \quad [4]$$

for a neutron fluence $nvt = 5.4 \times 10^{15}/\text{cm}^2$, from analysis of tritium release experiments. Evaluation of D at 450 C with Eq. [3] gives $1.7 \times 10^{-17} \text{ cm}^2/\text{s}$ and with Eq. [4] gives $6.1 \times 10^{-10} \text{ cm}^2/\text{s}$, a significant discrepancy. For a grain size R of 1 μm , $R \gg 2\sqrt{Dt}$ for the first value of D , and $R \approx 2\sqrt{Dt}$ for the second. The qualitative nature of the tritium build-up in the breeder materials can therefore not be determined using the available estimates of tritium diffusion coefficients. Upper limits of the diffusive inventory can be obtained, however, using the smaller values and solving Eqs. [1] and [2] over the particle residence time in the fluidized bed.

9.3.2 Low Temperature Tritium Release Mechanisms

For the range of parameters of interest, the tritium concentration within the grains builds during the pulse on-time and remains essentially unchanged during the off-time. Because tritium diffusion is slow relative to the pulse cycle, the tritium inventory builds in a manner similar to that in a

Figure 9-8. (a) Schematic of Tritium Concentration vs Time for $R < 2\sqrt{D\sigma}$
 (b) Schematic of Tritium Concentration vs Time for $R > 2\sqrt{D\sigma}$

continuous neutron flux, with total pulse on-time in the pulsed mode equivalent to total time in the steady-irradiation mode. Recoil ejection and knock-out, two low-temperature tritium release mechanisms that were not considered in the ANL STARFIRE analysis, were identified as potentially important in the fluidized bed. A model for recoil ejection was developed, and the fraction of tritium lost from grain clusters of various sizes by the recoil mechanism was calculated. Details are discussed below. For clusters from 0.1 to 1.0 mm in radius, which is the range of sizes proposed for the fluidized bed, ejection occurs from the near-surface region and involves on the order of ~1 to 25% of the total tritium produced. Although loss of this amount of tritium from the clusters reduces the inventory, it does not significantly alter the conclusion of the diffusive inventory calculation. The Knockout mechanism, which generally involves relatively low energy transfers from recoiling tritium or lattice atoms to a stationary tritium atom, is more of a "skin" effect than recoil and therefore affects a smaller, essentially negligible fraction of the gas.

RECOIL EJECTION

At low temperatures tritium is released from Li_2O primarily by temperature independent processes such as direct recoil or knock-out rather than by bulk diffusion, which dominates at high temperatures. A number of investigations of tritium release due to the recoil process have been performed with single crystals and sintered clusters of Li_2O at temperatures below 300°C .^(5,6) The design temperature range for the fluidized bed is 300° to 475°C , which lies below that anticipated in the STARFIRE design. Because of the relatively slow diffusion in this temperature range, it is desirable to determine if recoil ejection is an important tritium release mechanism for the cluster sizes proposed.

If we assume spherical clusters with uniform tritium production occurring throughout, the probability $P(r)$ that a triton recoil will lead to ejection depends on the recoil energy, the range of ^3H in the lithium compound of interest, and the size of the cluster. If r_T is the range of the triton and R is the cluster radius, this probability is given by

$$\begin{aligned}
 P(r) &= 0 \text{ if } r \leq R - r_T \\
 &= [(r_T + r)^2 - R^2]/4r_T r, \text{ if } r \geq R - r_T
 \end{aligned}
 \quad [1]$$

where r is the distance from the cluster center, and isotropic recoil has been assumed. $P(r)$ has been evaluated as a function of r for several values of cluster radius R . The results are plotted using the normalized distance r/R in Figure 9-9. The cross-hatched area indicates the pellet size range for the fluidized bed design. The range of the triton was taken as $38.4 \mu\text{m}^{(3)}$ for these calculations. For clusters with $R = 0.1 \text{ mm}$, recoil ejection is possible if $(r/R) \gtrsim 0.62$; for clusters with $R = 1 \text{ mm}$, if $(r/R) \gtrsim 0.96$. In the latter case, the outer "skin" is the only area contributing to release.

The tritium production rate $\dot{N}_p$ in a cluster of radius R is

$$\dot{N}_p = \frac{4}{3} \pi R^3 \dot{n} \quad [2]$$

where $\dot{n}$ is the production rate per unit volume per unit time. The recoil ejection rate is

$$\dot{N}_r = \int_0^R \int_0^\pi \int_0^{2\pi} \dot{n} P(r) r^2 \sin \theta \, d\theta \, d\varphi \, dr \quad [3]$$

and, using Equation [1], the loss fraction f_L is

$$f_L = \frac{\dot{N}_r}{\dot{N}_p} = \frac{3}{8} \left\{ -\frac{1}{6} (r/R)^3 - (r/R)^2 + \frac{3}{2} (r/R) - 1 \right\}. \quad [4]$$

This is plotted in Figure 9-10 for a range of cluster sizes R . As before, the cross-hatched area indicates the design range. From the figure, it is clear that only a small percentage of the tritium produced will be lost by the recoil process. The total tritium inventory in the bed will depend, therefore, on the bulk diffusion behavior within the grains.

Figure 9-9. Probability $P(r)$ that a Triton Recoil Will Lead to Ejection from a Li_2O Cluster of Radius R as a Function of r , Distance from Cluster Center.

Figure 9-10. Fraction of Tritons Lost From the Cluster, f_L , as a Function of Cluster Radius, R

Tritium release by the "knock-out" process requires energy transfer between recoiling lattice atoms or tritons and a stationary triton. The amount of energy transferred is generally smaller than the recoil energy; hence, a smaller fraction of the pellet volume contributes to release by this mechanism. In addition, the probability of this secondary energy transfer occurring depends on the local tritium concentration. The fraction of total tritium produced that is lost by knock-out can thus be neglected.

9.3.3 Conclusion

The implications of these results for the fluidized-bed design are straightforward. No significant improvement in the tritium release rate by the bulk diffusion mechanism is to be expected in the fluidized bed because of the low operating temperature (relative to the STARFIRE bed), and the inventory build-up, when this process is limiting, will occur at roughly the same rate as in a static packed bed at the same temperature. The recoil ejection mechanism, which does increase the total release rate, does not reduce the inventory by large amounts. Tritium removal must therefore be accomplished either by increasing the operating temperature or by removing the pellets, which can be done by adjusting the helium flow rate.

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