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AEC Research and Development Report

THE PACIFIC NORTHWEST LABORATORY
ANNUAL CONTROLLED THERMONUCLEAR
REACTOR TECHNOLOGY REPORT-1971

June 30, 1971

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THE PACIFIC NORTHWEST LABORATORY
ANNUAL CONTROLLED THERMONUCLEAR
REACTOR TECHNOLOGY REPORT - 1971

By

The Pacific Northwest Laboratory
Controlled Thermonuclear Reactor
Technology Committee

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June 30, 1971

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THE PACIFIC NORTHWEST LABORATORY
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I. INTRODUCTION

This report summarizes the research and development efforts of the Pacific Northwest Laboratories (PNL) for the advancement of Controlled Thermonuclear Reactor (CTR) technology. All CTR research conducted at PNL in FY71 was in the areas of engineering and technology. It is felt that there are a number of technological problems associated with the CTR power plant, exclusive of the plasma confinement problem, which could prove just as limiting to the progress of the total effort. PNL has identified and initiated research programs in certain of these areas.

Design of efficient CTR blankets or moderating regions is important to the development of the total power plant. Several CTR concepts involve the extraction of energy from neutrons emitted by the plasma. In addition, one of these concepts envisions the breeding of tritium from lithium by means of neutron capture such that the fusion reaction can be economically fueled. These two functions, energy extraction and breeding of tritium, must take place in a geometrically confined space. In addition, shielding must be provided for the superconducting region surrounding the blanket region of the CTR. In order to design such blanket regions to meet pre-determined geometrical, heat flux, shielding, and breeding ratio specifications, one must have a detailed knowledge of the physics phenomena involved when high energy (14.1 MeV and 2.45 MeV) neutrons interact with component materials at elevated temperatures. It is probable that the presently available fission neutron physics technology is applicable to CTR design to some degree. Indeed, some preliminary

design work is being carried out on blanket configurations with fission neutron design techniques. PNL is conducting neutronics research with two basic goals in mind. One goal is to determine the applicability of present fission neutron physics technology to the CTR program. The other goal is to develop, as necessary, the technology to supplement existing neutron physics knowledge required to make confident decisions related to CTR blanket design configurations. The status of these research efforts is reported in this document.

The PNL research program in CTR neutron physics involves experimental and calculational work initially aimed at determining the accuracy of present day blanket design techniques. An initial experiment, which was completed this year, involves the measurement of the reaction rate of 14 MeV incident neutrons in graphite at room temperature. A theoretical study, also completed this year, evaluates the accuracy with which two existing neutron physics computer codes can calculate the experimental data.

A second area of major concern in the design of CTR power plants is the magnitude of the neutron damage postulated to occur within the plasma chamber vacuum wall from the anticipated high neutron fluxes associated with certain CTR concepts. This area is being studied at PNL by simulation of this neutron damage phenomena with heavy ion bombardment. Direct study of the problem is difficult due to the absence of a high intensity, high energy, neutron source. Since over 90% of the damage from neutron bombardment occurs from the primary knock-on ion, neutron simulation of damage by ion bombardment appears to be the most attractive currently available

approach to damage simulation studies. The studies carried out this year include the determination of the material swelling rate as a function of temperature and fluence for niobium, molybdenum, and vanadium which are candidate materials for the primary vessel wall.

A CTR power plant of continuing interest to PNL is the fusion-fission concept. Analytical studies have been performed to determine energy multiplication, heat deposition, and breeding ratios as a function of geometry for a natural uranium, graphite, gas cooled lattice. The work completed this year is summarized.

One possible means of solving the storage problem associated with the long lived fission products created by fission power plants is to transmute them to more innocuous isotopes by neutron absorption. The feasibility of such a technique is dependent upon the availability of an intense source of neutrons. An initial study performed by PNL indicates that a CTR device may be the only possible candidate for providing such a neutron source. Work on this concept has been performed and is reported here.

A facility for efficiently carrying out CTR neutron physics experiments at operating temperatures will be required for the CTR technology program. A study has been performed on the desirability and applicability of converting the High Temperature Lattice Test Reactor (HTLTR) facility to this role. The results of this study are reported here.

Another task which has been initiated this year and which will be continued, is providing an up to date fit of theoretical functions to the fusion reaction cross section measurement. In the past, PNL has developed, through other research programs, what is perhaps the most powerful least squares fitting technique currently available. This technique is now being applied to the fitting of this cross section data. The work is

reported in this document.

Many of the CTR power plant concepts involve significant inventories of tritium and hydrogen. The effect of hydrogen on candidate CTR materials at expected operating temperatures is of interest to CTR designers. The results of an initial study in the area of concern is reported.

II. SUMMARY

Several research and development efforts have been undertaken at PNL during FY71 all of which contribute to the advancement of CTR technology. Some of the work reported here was supported on internal funds and other portions of the work was supported by related research programs.

The first of a series of planned bench mark experiments in 14 MeV neutron moderation in candidate CTR blanket materials was conceived, designed, and carried out. Absolute reaction rates from eleven reactions from 14 MeV incident neutrons covering the neutron spectrum from 14 MeV down to thermal were collected for a test assembly of room temperature graphite. The collected data was reduced and analyzed so as to place the information in a form suitable for evaluation of theoretical methods.

Calculations to compare the experimental results with theory were performed with two different transport theory codes. The results of this comparison result in several conclusions. They are:

- Both computer codes consistently underpredict the reaction rates as a function of penetration distance in the graphite.
- The two dimensional code underpredicts the reaction rates and the one-dimensional code overpredicts the reaction rates in the resonance and thermal range.
- The calculational envelope ranges over a factor of 10.

A fusion-fission parametric study was performed on a uranium graphite lattice. Several conclusions can be drawn from this study. First, if a depleted uranium converter is placed between the neutron source and the lattice, this converter makes a significant contribution to the fissile conversion rate. Secondly, over 99% of the thermal neutrons leaking out of the lattice reflector are absorbed in the first 10 cm of the following

lithium region. Finally, the thermal flux and therefore, the fission power rate in the fission blanket appears to have a peak average ratio of a factor of two or more.

The status of neutronics analysis of controlled thermonuclear blankets at PNL is reviewed in this document with a summary of the accomplishments to date. This is followed by a study of the CTR as a neutron source for nuclear transformation of fission products. This study led to the conclusion that at this time, only a CTR device offers the potential of a sufficiently large excess of neutrons to perform such a mission.

An extensive study of the applicability of the High Temperature Lattice Test Reactor (HTLTR) as a experimental facility for CTR blanket studies was performed. The approach is to use the facility as a large instrumented experimental cavity capable of being heated to temperatures up to 1100°C with the fission fuel removed. The conclusion of this study is that the HTLTR has the potential for being a primary facility for neutronic and other technical studies of blankets for CTR devices. In order to perform this role, however, several facility modifications are required.

A literature survey on neutron damage to superconductor materials was performed. One of the conclusions of this study is that meaningful irradiation experiments could be performed on candidate materials at a location two to three feet above the core of the Fast Flux Test Facility (FFTF).

In order to perform CTR engineering analysis studies at PNL, a basic CTR nuclear data file is being developed. The initial work involves non linear least squares fitting of some of the relative cross section

measurements to theoretical functional forms. Preliminary results indicate that the available data for $T(d,n)^4\text{He}$ can be described in a five parameter fit and the $D(d,p)T$ reaction can be described with a four parameter fit.

A study on the reaction of hydrogen and deuterium with niobium-zirconium alloys at 900°C was performed. The present study does not indicate any unusually high temperature absorption or desorption kinetics of hydrogen or deuterium by niobium alloys caused by the presence of zirconium. It is certain, however, that differences in reaction kinetics for niobium will be caused by the presence of alloy elements such as zirconium or yttrium.

Finally, a study was performed on radiation induced swelling of CTR materials by means of heavy ion bombardment techniques. The results of transmission electron microscopic investigations of exposed fusion reactor materials has revealed that swelling values of up to 1% will be produced in less than 1 month of exposure to future CTR radiation environments. It has also shown that inert gas atoms such as helium are not essential to the nucleation of voids. However, the inclusion of large amounts of neutronically produced helium and hydrogen in metals may increase the severity of the swelling problem.

III. DISCUSSION

This discussion consists of several individual papers reporting on several different pieces of work by many contributing authors. The work reported on includes efforts in neutronics, neutron damage simulation, superconductor damage, thermonuclear cross section analysis and reaction rates of hydrogen isotopes in niobium alloys. The specific papers are written in such a way as to be reasonably self contained.

A. Reaction Rates from 14 MeV Neutrons for Selected Foils in Room Temperature Graphite - L. D. Williams and D. F. Newman

Neutron flux and spectrum information has been obtained for ${}^3\text{H}(\text{d},\text{n}){}^4\text{He}$ source neutrons as a function of position in a pile of graphite at 20°C . Eleven reaction rates were measured using seven types of foils. These measurements provide a set of benchmark data to assess the accuracy of calculational methods planned for CTR blanket design.

The experiment also provided useful information on experimental techniques and neutron detectors which will be used when performing experiments with more complex blanket arrangements and at temperatures up to 1000°C .

Summary

Absolute reaction rates were determined at the test locations for each of the eleven reactions considered. Reduction and analysis of the counting and irradiation data was aimed at placing the experimental results in a form suitable for evaluation of theoretical methods. The experiment yielded results which are accurate enough to be used to define a benchmark problem for CTR blanket neutronics analysis methods.

Experimental Procedure

The experimental arrangement including the test assembly, the position of the neutron source, and the activation foils is shown in Figures A.1 and A.2.

The 37-1/2 x 37-1/2 x 45 in. deep test assembly consisted of criss-cross layers of GBF graphite bars, 3-3/4 x 3-3/4 x 37-1/2 in. in size. The assembly was stacked on a reinforced aluminum floor over a large concrete sump. The reactor grade graphite had a carbon density of 1.6714 gm/cm³ and a 2200 m/sec absorption cross section of 3.80 ± 0.04 mb including all impurities.⁽¹⁾ Nitrogen and oxygen densities in the bars were 2.654×10^{-4} and 8.133×10^{-5} gm/cm³, respectively. The center graphite bar in the test assembly was removable. Special graphite foil holders shown in Figure A-3 fit into slots in the graphite bar.

The irradiation was conducted in a positive ion accelerator target room which was designed to be scatter-free for neutrons. The neutron source was provided by a neutron generator with an accelerating voltage of about 95 keV per deuteron. The source of 14 MeV neutrons had an average strength during the irradiation of about 7×10^{10} neutrons per second.

Seven different types of foils were used in the experiment. They were held in the special foil holders by 1 mil mylar tape. Activities from eleven nuclear reactions were obtained from the foils. The foil specifications and the reactions of interest are summarized in Table A.1.

The foils were irradiated for 161 minutes. Neutron fluence and flux data were obtained by two precision long counters positioned at calibrated locations from the source. A printout of one of the counters every minute provided information on the stability of the source output during the irradiation.

Following the irradiation, the foils were removed from the test assembly and their activity counted. The activity of several foils of each type were absolutely determined using a calibrated GeLi detector and associated equipment. Relative activities were determined for each type of foil by integral counting techniques using a NaI(Tl) detector.

FIGURE A.1. Experimental Arrangement

FIGURE A.2. Graphite Test Assembly

FIGURE A.3. Foil Holder with Foils

The relative activities from integral counting data were converted to absolute values by normalizing to the foils which were counted on the GeLi system. The photopeaks used for the reaction product analysis and the energy discrimination levels used during integral counting are shown in Table A.2.

The total counts under the photopeak of interest for a counting interval following the irradiation were translated into foil activities at shutdown by the following equation.

$$A_o = \frac{\Delta t}{LT} \cdot \frac{C\lambda}{(1-e^{-\lambda\Delta t}) e^{-\lambda t_1}}$$

where

A_o = foil activity, counts per unit time per foil

Δt = counting interval, clock time

LT = counting interval, live time

C = total counts under photo peak of interest (less background) obtained during the counting interval

λ = decay constant of reaction product

t_1 = time from irradiation shutdown to start of count.

The source output was not constant during the irradiation period. However, investigations of the minor variations revealed that the standard equation which assumes a constant neutron flux was adequate (< 1%) to describe the saturated activity for all of the reaction products. Thus, the foil activities were converted to saturated activities (infinite irradiation period) by

$$A_{SAT} = A_o / (1 - e^{-\lambda\Delta T})$$

TABLE A.1

FOIL SPECIFICATIONS AND NUCLEAR REACTIONS

<u>Foil</u>	<u>Diameter, inches</u>	<u>Thickness, inches</u>	<u>Average Foil Weight, grams</u>	<u>Reactions</u>
Au	0.5	0.0005	0.0328	$^{197}\text{Au}(n,\gamma)^{198}\text{Au}$ $^{197}\text{Au}(n,2n)^{196}\text{Au}$
Cu	0.5	0.005	0.1522	$^{63}\text{Cu}(n,\gamma)^{64}\text{Cu}$ $^{65}\text{Cu}(n,2n)^{64}\text{Cu}$
Mg	0.5	0.005	0.0300	$^{24}\text{Mg}(n,p)^{24}\text{Na}$
Al	0.5	0.250	2.1950	$^{27}\text{Al}(n,p)^{27}\text{Mg}$ $^{27}\text{Al}(n,\alpha)^{24}\text{Na}$
80% Mn-Cu	0.5	0.002	0.0470	$^{55}\text{Mn}(n,\gamma)^{56}\text{Mn}$
Ti	0.5	0.010	0.1585	$^{48}\text{Ti}(n,p)^{48}\text{Sc}$
Ni	0.5	0.010	0.2815	$^{58}\text{Ni}(n,p)^{58}\text{Co}$ $^{58}\text{Ni}(n,2n)^{57}\text{Ni}$

TABLE A.2

SUMMARY OF VALUES USED DURING COUNTING AND DATA REDUCTION FOR VARIOUS REACTION PRODUCTS

Foil	Reaction	GeLi System		NaI(Tl) System		Reaction Product Half Life
		Gamma-Ray Used in Analysis, keV	Gamma-Ray Intensity, %	Energy Discrimination Level, keV		
Au	$^{197}\text{Au}(n,\gamma)^{198}\text{Au}$	412	95	380		2.697
	$^{197}\text{Au}(n,2n)^{196}\text{Au}$	333, 356	25, 94	NA ⁽¹⁾		6.18
Cu	$^{63}\text{Cu}(n,\gamma)^{64}\text{Cu}$	511 ⁽²⁾	38	430 ⁽²⁾		12.80 hr
	$^{65}\text{Cu}(n,2n)^{64}\text{Cu}$					
Mg	$^{24}\text{Mg}(n,p)^{24}\text{Na}$	1369	100	1000		15.0 hr
Al	$^{27}\text{Al}(n,p)^{27}\text{Mg}$	840	70	NA		9.5 min
	$^{27}\text{Al}(n,\alpha)^{24}\text{Na}$	1369	100	1000		15.0 hr
80% Mn-Cu	$^{55}\text{Mn}(n,\gamma)^{56}\text{Mn}$	847	99	760		2.58 hr
Ti	$^{48}\text{Ti}(n,p)^{48}\text{Sc}$	983, 1040, 1314	100, 100, 100	940		1.83
Ni	$^{58}\text{Ni}(n,p)^{58}\text{Co}$	810 ⁽³⁾	99	730 ⁽³⁾		71.3
	$^{58}\text{Ni}(n,2n)^{57}\text{Ni}$	1370	86	1000		36.0 hr

(1) Na - Not Applicable, Reaction Product Was **Not** Counted Using This System

(2) Both Cu Reactions Yielded The Same Product

(3) Reaction Product Was Counted Two Weeks Following Irradiation To Allow For Decay Of 36 hr ^{57}Ni

where

A_{SAT} = saturated activity, counts per unit time per foil

ΔT = duration of irradiation, 161 minutes

λ = decay constant of reaction product of interest.

Absolute reaction rates listed in Table A.3 and graphically displayed in Figure A.4, were calculated from the saturated foil activities by applying counter efficiency and gamma-ray intensity factors⁽²⁾ and by normalizing to unit target nuclei as shown below.

$$R = \frac{A_{SAT}}{\epsilon IN}$$

where

R = reaction rate, reactions per unit time per target nuclei

ϵ = detector efficiency, counts per gamma-ray emitted

I = gamma-ray intensity, gamma-ray per disintegration of reaction product

N = total number of target nuclei per foil.

The relative reaction rate distributions in the test assembly were determined from integral counting data. The raw counting data was reduced to relative activities by using the computer code APDAC.⁽³⁾

Since it was experimentally impossible to discern which copper isotope yielded ^{64}Cu , the reaction rates for the two Cu reactions are given per Cu nuclei rather than per target nuclei.

Uncertainties in the reported reaction rates were derived from counting statistics. Uncertainties in detector efficiencies, gamma-ray intensities, half-lives, and foil weights were assumed to be negligible compared to the counting statistics. As indicated in Table A.3, factors for self shielding of target nuclei have not been applied to the reaction rates at

TABLE A.3

FOIL ACTIVATION RESULTS

Reaction	Reaction Rate $\times 10^{20}$		Reaction/Sec Per Target Nuclei ⁽¹⁾							
	Position 0	Position 1	Position 2	Position 3	Position 4	Position 5	Position 6	Position 7	Position 8	Position 9
$^{27}\text{Al}(n,p)^{27}\text{Mg}$	1233 ± 6	91.3 ± 2.6	----	5.9 ± 0.3						
$^{27}\text{Al}(n,\alpha)^{24}\text{Na}$	1926.5 ± 8.3	101.1 ± 1.8	18.2 ± 0.3	4.65 ± 0.09	1.41 ± 0.06	0.52 ± 0.02	----	0.14 ± 0.02	----	0.08 ± 0.02
$^{24}\text{Mg}(n,p)^{24}\text{Na}$	4071 ± 65	170.0 ± 3.2	31.6 ± 1.0	9.9 ± 0.8	4.48 ± 0.86	2.89 ± 0.91	----	1.14 ± 0.87	----	----
$^{55}\text{Mn}(n,\gamma)^{56}\text{Mn}$	7715 ± 69	24512 ± 245	31049 ± 310	28925 ± 289	22975 ± 230	16065 ± 161	10329 ± 103	6090 ± 61	2997.5 ± 30	648.3 ± 7.8
$^{197}\text{Au}(n,2n)^{196}\text{Au}$	70803 ± 991	4845 ± 591	1770 ± 602							
$^{197}\text{Au}(n,\gamma)^{198}\text{Au}$	38179 ± 447	125752 ± 817	151216 ± 983	127995 ± 1050	90724 ± 771	59166 ± 521	35985 ± 324	20417 ± 204	9920 ± 129	2065 ± 74
$^{63}\text{Cu}(n,\gamma)^{64}\text{Cu}$	$8760 \pm 51^{(2)}$	$2252 \pm 29^{(2)}$	$2454 \pm 32^{(2)}$	$2270 \pm 27^{(2)}$	$1794 \pm 23^{(2)}$	$1268 \pm 17^{(2)}$	$808 \pm 11^{(2)}$	$481 \pm 8^{(2)}$	$235 \pm 5^{(2)}$	$51 \pm 3^{(2)}$
$^{65}\text{Cu}(n,2n)^{64}\text{Cu}$										
$^{58}\text{Ni}(n,2n)^{57}\text{Ni}$	678.0 ± 23.1	22.2 ± 1.6	5.79 ± 1.27	2.05 ± 1.44						
$^{58}\text{Ni}(n,p)^{58}\text{Co}$	8225 ± 82	569 ± 80	152 ± 57							
$^{48}\text{Ti}(n,p)^{48}\text{Sc}$	1275 ± 20	41.3 ± 1.6	8.2 ± 0.9	2.4 ± 1.0						

(1) Factors For Self Shielding of Target Nuclei Have Not Been Applied

(2) Since Both Reactions Yield the Same Product, The Reaction Rate Per Cu Nuclei is Presented Rather Than Per Target Nucleus.

FIGURE A.4. Reaction Rates for Various Target Nuclei as a Function of Position in a Graphite Pile Using a 14 MeV Neutron Source

this stage of the analysis.

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- B. A Calculation-Experiment Comparison of 14 MeV Neutrons Slowing Down in Graphite - L. D. Williams, U. P. Jenquin, B. R. Leonard, W. C. Wolkenhauer, and D. F. Newman

The bulk of the useful energy in most CTR concepts is available in the form of high energy neutrons. Specifically, one of the most promising reactions (deuterium-tritium) gives off most of its energy in the form of a 14.1 MeV neutron. This energy is extracted in a CTR design by moderation of the neutrons in a blanket region surrounding the plasma. In addition, in a D-T CTR the thermalized neutrons must be used to breed tritium from lithium. Design calculations for these types of systems have been performed by several investigators.^(4,5,6) The calculations are usually performed using transport theory computer codes developed under fission reactor programs. In some cases, Monte Carlo techniques have been employed. The accuracy of these computational methods must be determined when they are applied to the higher energy neutrons associated with the CTR systems.

The question of the adequacy of currently available neutron transport codes for CTR blanket design calculations is of more than academic interest. CTR blanket designers will have to consider several major criteria in doing conceptual design. In order to meet any design requirements based on these considerations, detailed knowledge of the slowing down phenomena of fusion neutrons in reactor blankets is essential. For a D-T CTR, one of the most important considerations is that the tritium breeding ratio be accurately predictable. For all blanket designs, the ability to accurately specify the proper blanket thickness is essential. In addition, accurate calculation of the power distribution and after-

heat production is required. All of these parameters depend upon knowledge of the cross sections and neutron flux as a function of energy and position. A misjudgement of the thermal neutron portion of the total neutron flux in a CTR blanket, for instance, can lead to a serious misprediction of the tritium breeding ratio.

To help in making an evaluation of the applicability of these methods, a comparison is made between the experimental results reported in the previous section and two representative transport theory codes. The data collected from the experiment consists of reaction rates for the various foils as a function of axial position in the graphite. Reaction rates for the detector materials were calculated by the transport theory codes and compared to the measured reaction rates.

Briefly, the experiment consisted of irradiating various neutron activation foils by a 14 MeV neutron source. The foils were selected so that the neutron energy spectrum was covered from 14 MeV to thermal energies. The various foils were located every five inches along the axis of a 45 inch thick block of reactor grade graphite. The experimental configuration including the position of the neutron source is shown in Figures A.1 and A.2.

The ANISN⁽⁸⁾ code and the TWOTRAN⁽⁹⁾ code were used to calculate the reaction rates. The former is a one dimensional multigroup transport theory code which has been used in several conceptual design calculations reported in the literature. The latter is a two dimensional multigroup transport theory code which has just recently become available. Calculations were performed with the ANISN code in both slab and spherical geometries. It

is hoped that these two geometries constitute an envelope of the capability of ANISN to calculate the reaction rates. Because of its two-dimensional capability, the TWOTRAN code provided a reasonable simulation of the experiment.

Summary and Conclusions

Calculations were performed with two different transport theory codes. The calculated reaction rates were compared to measured reaction rates in order to determine the applicability of these codes to CTR blanket design. A number of conclusions can be drawn from the comparison. They include:

- Both ANISN and TWOTRAN codes consistently under predict the reaction rates as a function of penetration distance in the graphite.
- TWOTRAN underpredicts the reaction rates and ANISN overpredicts the reaction rates in the resonance and thermal energy range.
- The calculational envelope ranges over a factor of 10.

Because of underprediction of the threshold reaction rates and underprediction of the thermal reaction rates, one can say that TWOTRAN would tend to underpredict the tritium breeding ratio. By the same token, ANISN would tend to overpredict the tritium breeding ratio. In general, one could conclude from this comparison that the calculational methods and cross sections currently available are not sufficiently accurate to make confident predictions as to the neutronics characteristics of CTR blankets.

Calculational Method

Transport theory calculations were made for an isotropically emitting source of 14.5 MeV neutrons incident on a graphite block. The majority of the 100 group cross sections are from ENDF/B Version-II which was obtained

from ORNL. ANISN was used to collapse the 100 group cross sections into 27 groups. The 100 group cross sections had a group structure the same as the GAM-II code. The 27 group structure is given in Table B.1.

Macroscopic carbon cross sections were used with anisotropic scattering up to P_3 . Microscopic activation cross sections were used for the foil reaction rate calculations. The cross sections for the thermal group were obtained from a THERMOS calculation in graphite at room temperature with no leakage.

Since some of the neutron cross sections were not included in the data obtained from ORNL, the cross section had to be obtained from other sources.

Data for the $^{197}\text{Au}(n,2n)$, and $^{27}\text{Al}(n,\alpha)$ reactions were taken from ENDF/B listings, $^{58}\text{Ni}(n,p)$ from Bresesti, and $^{24}\text{Mg}(n,p)$, $^{27}\text{Al}(n,p)$, $^{65}\text{Cu}(n,2n)$, $^{48}\text{Ti}(n,p)$, and $^{58}\text{Ni}(n,2n)$ from an evaluation by B. R. Leonard. Using this data, 27 group cross sections were developed.

The activation cross sections in the top energy group (from 13.5 to 14.5 MeV) were increased by a factor of 1.431 because the source neutrons were assumed to have a discrete energy of 14.5 MeV. Since the incident neutron lethargy is -0.3716, it takes longer for the neutrons to get out of this group than the calculation indicates. Therefore, the actual flux is higher than the calculated flux by a factor of

$$(-.3716 + .3000)/\frac{(-.4 + .3)}{2} = 1.431$$

Thus, to get the right reaction rates, the group 1 cross section were increased by a factor of 1.431.

TABLE B.1

GROUP STRUCTURE USED IN CTR TWOTRAN AND ANISN CALCULATIONS

<u>Group No.</u>	<u>Energy Interval, eV</u>		<u>Lethargy Interval</u>	
1	1.4918247 + 07	to 1.3498588 + 07	-0.40	to -0.30
2	1.3498 + 07	1.2214 + 07	-0.30	-0.20
3	1.2214 + 07	1.1052 + 07	-0.20	-0.10
4	1.1052 + 07	1.0000 + 07	-0.10	0.00
5	1.0000 + 07	9.0484 + 06	0.00	0.10
6	9.0484 + 06	8.1873 + 06	0.10	0.20
7	8.1873 + 06	7.4082 + 06	0.20	0.30
8	7.4082 + 06	6.7032 + 06	0.30	0.40
9	6.7032 + 06	6.0653 + 06	0.40	0.50
10	6.0653 + 06	5.4881 + 06	0.50	0.60
11	5.4881 + 06	4.4933 + 06	0.60	0.80
12	4.4933 + 06	3.6788 + 06	0.80	1.00
13	3.3768 + 06	3.0119 + 06	1.00	1.20
14	3.0119 + 06	3.4660 + 06	1.20	1.40
15	2.4660 + 06	2.0190 + 06	1.40	1.60
16	2.0190 + 06	1.1080 + 06	1.60	2.20
17	1.1080 + 06	6.0810 + 05	2.20	2.80
18	6.0810 + 05	2.7324 + 05	2.80	3.60
19	2.7324 + 05	1.1109 + 05	3.60	4.50
20	1.1109 + 05	4.0868 + 04	4.50	5.50
21	4.0868 + 04	9.1188 + 03	5.50	7.00
22	9.1188 + 03	1.2341 + 03	7.00	9.00
23	1.2341 + 03	1.6702 + 02	9.00	11.00
24	1.6702 + 02	2.2603 + 01	11.00	13.00
25	2.2603 + 01	3.0590	13.00	15.00
26	3.0590	0.4140	15.00	17.00
27	0.4140	0.0	17.00	0.0

The 27 group macroscopic carbon cross sections and the microscopic activation cross sections were put onto a tape in a format to use with ANISN. From there they were put onto another tape in TWOTRAN format with the $2\ell + 1$ factor removed from the Legendre expansion of the scattering matrix.

The graphite pile was represented in the TWOTRAN calculation as a right cylinder with a radius of 21.157 in. and a height of 46.25 in. There was 9 radial intervals and 21 axial intervals. The first axial interval which represents air is 1.102 in. thick. The intervals were selected such that the center of the interval was at the foils position. The first foil position was 0.25 in. from the front edge of the graphite. The source was put in as a bottom boundary source for the first radial interval. The calculation was done with S_8 quadratures.

Two ANISN calculations were made for the experiment, neither of which comes very close to representing the actual system. The first was a slab calculation with a graphite thickness of 45.5 in. and zero flux boundaries. There was no perpendicular leakage. The source was a plane source at the edge of the graphite. The spherical calculation was done with a 2.2 in. radius central void containing the source. The graphite was 45.5 in. thick. The ANISN calculations used the same cross sections as the TWOTRAN calculation. They were S_8 calculations with P_3 cross sections.

Since each reaction rate obtained from the calculations is a summation of the group cross sections times the fluxes calculated at each space point, the activities represents saturated, infinitely dilute reaction rates per target nucleus. The experimental results have been reported in the form of saturated reaction rates per target nuclei. In other words, the experimental data has not been corrected to dilute reaction rates. The outer flux

perturbation and the self shielding corrections for each type of detector vary as a function of the neutron spectrum. For this reason, the infinitely dilute activities obtained from each calculation were corrected by using the calculated spectrum and multigroup cross section for each reaction. The self shielding and foil perturbation corrections were calculated using prescriptions given in Reference 10.

Eleven reaction rates were calculated. The reactions and their appropriate energy ranges are shown in Table B.2. The reactions are listed in

TABLE B.2
THE REACTIONS AND THEIR ENERGY RANGE

<u>Reaction</u>	<u>Energy Range</u>
$^{58}\text{Ni}(n,2n)^{57}\text{Ni}$	$E > 12 \text{ MeV}$
$^{65}\text{Cu}(n,2n)^{64}\text{Cu}$	$E > 10 \text{ MeV}$
$^{197}\text{Au}(n,2n)^{196}\text{Au}$	$E > 8 \text{ MeV}$
$^{27}\text{Al}(n,\alpha)^{24}\text{Na}$	$E > 6 \text{ MeV}$
$^{24}\text{Mg}(n,p)^{24}\text{Na}$	$E > 5 \text{ MeV}$
$^{48}\text{Ti}(n,p)^{48}\text{Sc}$	$E > 3 \text{ MeV}$
$^{27}\text{Al}(n,p)^{27}\text{Mg}$	$E > 2 \text{ MeV}$
$^{58}\text{Ni}(n,p)^{58}\text{Co}$	$E > 1 \text{ MeV}$
$^{197}\text{Au}(n,\gamma)^{198}\text{Au}$	Resonance + Thermal
$^{63}\text{Cu}(n,\gamma)^{64}\text{Cu}$	Resonance + Thermal
$^{55}\text{Mn}(n,\gamma)^{56}\text{Mn}$	Resonance + Thermal

the table in order of their neutron energy coverage from fast to thermal.

The measured and corrected calculated reaction rates are shown in Figures B.1 through B.10. The figures are in the same order as the reactions listed in Table B.2 except that the Cu reactions are combined in one figure. A consistent underprediction of the reaction rates can be noticed from high energy neutrons as a function of penetration distance in the graphite. A very wide calculational envelope in the resonance and thermal region is also apparent. In general, the calculation under-predicts the neutron penetration and over-predicts the slowing down rate.

FIGURE B.1. Reaction Rates for ^{58}Ni (n,2n) ^{57}Ni as a Function of Position in a Graphite Pile Using a 14 MeV Neutron Source

FIGURE B.2. Reaction Rates for $^{197}\text{Au}(n,2n)^{196}\text{Au}$ as a Function of Position in a Graphite Pile Using a 14 MeV Neutron Source

FIGURE B.3. Reaction Rates for $^{27}\text{Al}(n,\alpha)^{24}\text{Na}$ as a Function of Position in a Graphite Pile Using a 14 MeV Neutron Source

FIGURE B.4. Reaction Rates for $^{24}\text{Mg}(n,p)^{24}\text{Na}$ as a Function of Position in a Graphite Pile Using a 14 MeV Neutron Source

FIGURE B.5. Reaction Rates for $^{48}\text{Ti}(n,p)^{48}\text{Sc}$ as a Function of Position in a Graphite Pile Using a 14 MeV Neutron Source

FIGURE B.6. Reaction Rates for $^{27}\text{Al}(n,p)^{27}\text{Mg}$ as a Function of Position in a Graphite Pile Using a 14 MeV Neutron Source

FIGURE B.7. Reaction Rates for $^{58}\text{Ni}(n,p)^{58}\text{Co}$ as a Function of Position in a Graphite Pile Using a 14 MeV Neutron Source

FIGURE B.8. Reaction Rates for $^{197}\text{Au}(n,\gamma)^{198}\text{Au}$ as a Function of Position in a Graphite Pile Using a 14 MeV Neutron Source

FIGURE B.9. Reaction Rates for $^{63}\text{Cu}(n,\gamma)^{64}\text{Cu}$ & $^{65}\text{Cu}(n,2n)^{64}\text{Cu}$ as a Function of Position in a Graphite Pile Using a 14 MeV Neutron Source

FIGURE B.10. Reaction Rates for $^{55}\text{Mn}(n,\gamma)^{56}\text{Mn}$ as a Function of Position in a Graphite Pile Using a 14 MeV Neutron Source

C. A Fusion-Fission Parametric Study - U. P. Jenquin, B. R. Leonard, Jr., D. H. Thomsen, and W. C. Wolkenhauer

Several previous investigations have studied the use of fissile or fertile material in CTR blankets in order to obtain enhanced power generation from fission and/or breeding of fissile material. Barrett⁽¹¹⁾ proposed a thermal fission reactor blanket which could be made critical for pre-heating or power generation in the absence of the CTR source. Lidsky⁽¹²⁾ studied the incorporation of fertile material in the blanket to provide fissile material to be utilized in a separate fission reactor. Lontai⁽¹³⁾ made a thorough study of the use of a thin uranium convertor region which provided a margin of neutron multiplication and an overall energy gain of about a factor of two. Lee⁽¹⁴⁾ has studied the neutronics properties of rather thick blanket systems of Th, natural U, and depleted U. These systems were found capable of producing, in principle, unit tritium breeding, an energy gain of approximately a factor of ten through conversion to fission energy, and conversion ratios for producing fissile from fertile material significantly greater than unity. Lee's results for natural uranium were in agreement with the results expected from the experiments of Weale, et.al⁽¹⁵⁾ who measured the activations due to DT neutrons in thick uranium blankets.

The objective of the fusion-fission studies reported here differs from that of previous work. This objective has been to characterize fission blanket systems which will provide maximum power multiplications of DT source neutrons subject to several restrictions:

- The fission blanket is basically a thermal reactor system.

- The fission blanket is clearly subcritical.
- Unit tritium breeding is obtained.
- Fissile material is conserved.

This paper reports the initial results of a parametric study of natural uranium, graphite, helium-cooled lattices. The results⁽¹⁶⁾ of a preliminary study of a beryllium-moderated blanket indicated that a graphite system would probably achieve the required specifications and, therefore, graphite has been chosen for the present study as a more practical material.

Conceptual Design

A subcritical thermal fission-reactor lattice with an effective multiplication constant k will produce a number of fissions N , per source neutron of;

$$N = \frac{1}{\nu} \frac{k}{1-k} .$$

For a value of $k = 0.9$ and an effective number of neutrons emitted per fission $\nu \approx 2.5$ such a system will provide about 3.5 fissions per source neutron. The total heat energy produced in the blanket by a 14 MeV neutron thermalized and absorbed to produce fission is approximately a factor of ten greater than that which would have been produced by ${}^6\text{Li}(n,\alpha)\text{T}$ absorption in lithium. Thus the energy multiplication of a fission blanket with $k = 0.9$ is a factor of about 35 except as might be modified by the requirement that at least one neutron must be absorbed in lithium for tritium production.

A simple thermal fission lattice of natural uranium, however, consumes fissile ${}^{235}\text{U}$ faster than ${}^{239}\text{Pu}$ is created from neutron capture in

^{238}U for values of k approaching unity. Thus, while such a system could provide an appreciable multiplication of DT plasma energy it would deplete the overall energy reserves.

As previously noted, however, a blanket of uranium surrounding a 14 MeV neutron source can provide both neutron multiplication and breeding of fissile material.^(14,15) In the present concept, therefore, we propose that the first cylindrical annulus contain a high concentration of uranium. This annulus will, hopefully, provide the desired excess breeding of fissile material and provide an enhanced source of neutrons for increased power multiplication in a surrounding annular subcritical fission lattice.

The Calculational Model

The CTR vacuum chamber was considered to be a cylinder of 100 cm radius. The only source calculation done to date used a uniform isotropic neutron source of 14.2 MeV average energy in a 20 cm radius. In future calculations we intend to extend the source to 80% of the vacuum wall radius.

The model of the CTR blanket consisted of four annular regions. These were:

1. A convertor region containing $5/12$ ^{238}U , $1/6$ Nb and $5/12$ void.
2. A homogeneous fission lattice corresponding to one inch natural uranium rods on an 8 inch pitch.
3. A graphite reflector-moderator, and
4. A natural lithium blanket consisting of 45% Li, 45% C, and 10% Nb.

The parameterization of the neutronic properties of the CTR blanket was obtained by adjusting the thicknesses of the four annular regions as indicated by calculated results.

The Calculational Method

The calculations were done with ANISN using a 27 energy group structure. In general, ENDF/B-II 100 group cross sections obtained from ORNL were used in the calculation. The 100 group cross sections for U-235, U-238, Nb, C, Li-6, and Li-7 were collapsed into 27 group cross sections with ANISN assuming a homogeneous system of 1 in. natural uranium rods in an 8 in. square graphite lattice with no leakage. Thermal cross sections were obtained with a THERMOS calculation of the natural uranium rod in the graphite lattice. The temperature was assumed to be 1000°C.

Since the cross sections punched from the ANISN collapsing calculation represent infinitely dilute reaction rates, resonance self-shielding is not included. To obtain the correct U-238 absorption cross sections, two HRG3 calculations were done. An HRG3 calculation for the uranium rod in the graphite lattice was done assuming a fuel temperature of 1200°K. Another HRG3 calculation was done with smooth cross section data. The ratio of cross sections for U-238 between the two cases was the correction factor applied to the ANISN cross sections.

The Nb (n,2n), and ^{238}U (n,2n) reactions are included only in the scattering matrix of the ANISN punched cross sections. Cross sections for these reactions were hand calculated based on ENDF/B Version-II data. In order to conserve neutrons, the absorption cross sections were reduced by the appropriate amount to account for (n,2n) and (n,3n) reactions in ^{238}U and (n,2n) reactions in Nb. The ANISN calculations were done in cylindrical geometry with an S_8 quadrature set. The fission blanket contained anisotropic cross sections up to P_3 . The other regions contained P_0 cross sections.

Reaction rates were obtained in the ANISN runs for U-238 absorption and fission, U-235 absorption and fission, and Li-6 absorption. From these

numbers one can obtain the total and spatial Pu-239 breeding ratios, tritium breeding ratio, and power generation.

The first series of calculations were performed to determine the effective fission neutron multiplication constant of the blanket without any 14 MeV source neutrons. These calculations serve to parametrize the neutronics of the blanket systems with the CTR shut down.

The second series of calculations were designed to study the neutronics properties of blankets which had values of $k \approx 0.9$ in the first series of calculations when the 14 MeV neutron source was included. In the source calculations with the convertor present, it was seen that the neutron flux spectra changed greatly from the spectra without the source and convergence of spectral shapes was very slow. Utilizing the diffusion theory option in ANISN improved the convergence rate but we have not yet obtained a convergence which we judge to give reliable results for the source calculations.

Discussion of the Results

The results of the first series of calculations with only a fission spectrum source are shown on Table C.1. In the first three cases only the thickness of the fission lattice was varied. The effective multiplication factor is seen to change significantly with this dimension. This effect was expected since the specifications chosen for the lattice would produce a multiplication factor greater than one for that lattice, by itself, of infinite dimensions.

Case 5 is identical to case 2 except that the reflector graphite thickness has been doubled. The increased reflector has a significant effect on multiplication, $\Delta k = + 0.032$. However, it should be noted that this

increase is at the expense of reduced leakage to the lithium region and the required leakage for tritium breeding has not yet been determined.

Case 4 is identical to case 2 except that the thickness of the convertor region has been decreased from 12 cm to 6 cm. The consequent effect on multiplication is small. Comparisons of cases 6 and 3 also indicate a small effect for the thicker fission lattice when the effects of doubling the reflector thickness are taken into account. The implication of this result is that the 6 cm thickness is sufficient to absorb a large fraction of the thermal and resonance energy neutrons. This comparison does not imply anything about the relative properties of the convertor thickness in the presence of the 14 MeV source. The results of case 7, in which the convertor has been replaced by fission lattice, also demonstrates the extent that the convertor depresses the neutron multiplication.

The lithium blanket was reduced to 20 cm in the later cases since it was determined that greater thicknesses contributed insignificantly to tritium breeding. This result says nothing, however, about the effectiveness of shielding against leakage from the blanket.

Source calculations were also made for case 7 without the convertor. Although complete convergence was not obtained the calculated results of multiplication constant and fission breeding ratio were within about 20 percent of the expected values.

Although the source calculations with the convertor have not yet yielded reliable quantitative results some important qualitative trends are apparent. Figure C.1 illustrates several of these for the source calculation corresponding to case 6 of Table C.1. First we observe that the convertor region is making a significant contribution to the fissile con-

version rate and preliminary results indicate that the conversion ratio is greater than unity. Secondly we note that over 99% of the thermal neutrons leaking from the reflector are absorbed in the first 10 cm of the lithium larger. Finally we note that the thermal flux and, hence, the fission power rate in the fission blanket appears to have a peak average ratio of a factor of two or more. Although this effect was anticipated in this simple design concept, it illustrates a potential problem in achieving a practical fission blanket design.

FIGURE C.1. Relative Thermal Flux Distribution

TABLE C.1
SUMMARY OF BLANKET MULTIPLICATION CALCULATIONS

<u>CASE NO.</u>	<u>CONVERTER THICKNESS cm</u>	<u>LATTICE THICKNESS cm</u>	<u>REFLECTOR THICKNESS cm</u>	<u>BREEDER THICKNESS cm</u>	<u>TOTAL BLANKET THICKNESS cm</u>	<u>NEUTRON MULTIPLICATION</u>
1	12	100	20	∞	∞	.688
2	12	140	20	30	202	.800
3	12	200	20	30	262	.893
4	6	140	20	30	196	.809
5	12	140	40	30	222	.832
6	6	200	40	20	266	.920
7	-	206	40	20	266	.969

D. The Status of Neutronic Analysis of Controlled Thermonuclear Reactor Blankets at PNL - B.R. Leonard, Jr.

An effort has begun at PNL to study the neutronic characteristics of CTR blankets. The accomplishments to date include:

1. A preliminary calculation of the power-multiplication and breeding characteristics of a natural-uranium convertor, BeO-natural U fission lattice driven by 14-MeV neutrons.
2. The preparation of an abstract, based on the above calculation, submitted for the June, 1971 IAEA fusion conference.
3. The preparation of a document, not yet released for external distribution, which discusses the status of fission-fusion systems and some of the technical problems to be faced in the analysis of such systems.
4. The completion of an experiment (see Section A) in which a graphite pile slab was irradiated by 14 MeV neutrons and the resultant axial neutron flux energy spectrum characterized by neutron activation detectors.
5. Making operational for analysis a two-dimensional, discrete-ordinates neutron transport code, TWOTRAN, for analysis of CTR blankets and experimental results from item 4.
6. Designating the eleven activation cross sections, and reevaluating five of these, to be used in comparing measured and calculated reaction rates in the graphite pile irradiation.
7. Define preliminary specifications of a natural U-graphite lattice for future CTR blanket analysis.

8. Study and make preliminary specifications of the modifications to the High-Temperature Lattice Test Reactor which are necessary for utilization of the facility for a long-range experimental program of CTR blanket analysis.
9. Make survey calculations of the feasibility of using a CTR for the nuclear transformation of fission products.
10. A document was prepared and issued describing the potential use of the Hanford plant site as a location for a CTR prototype plant.

A credible analysis of a CTR blanket driven, at least in part, by 14 MeV neutrons, will require careful attention both to the nuclear data used in the analysis and to the calculational methods employed. Not only are credible data and methods to be required but the uncertainties in calculated quantities introduced by uncertainties in the data and approximations used in the calculational methods must be quantitatively defined.

In planning the calculational methods to be used, the assumption was made that neutron diffusion theory methods will be generally inadequate. However, their utilization for survey calculations are planned for study in the future because of the lesser expense involved in their use. The one-dimensional transport code ANISN, which has been widely used in CTR blanket analysis has been evaluated and is routinely in use at PNL. The two-dimensional transport code TWOTRAN had been assembled at PNL but not severely tested. The characteristics of the code and limitations of energy and spatial complexes have been studied in terms of anticipated problems of 14 MeV neutron sources and the code is now operational for this analysis. The Battelle Monte Carlo code, BMC, has been operational for some time. The effective utilization of the Monte Carlo method

requires the results of other, less expensive, calculations to aid in the convergence of the Monte Carlo routines. We plan, in the future, to recast BMC for application to this problem when results from transport calculations have been obtained and studied. The importance of careful consideration to calculational methods is justified, e.g., in the disparity of results obtained by Lee⁽¹⁷⁾ using transport and Monte Carlo methods.

As previously discussed, the nuclear data specifications are equally important to the calculational methods employed. To this end, PNL has given some consideration to Niobium cross sections. Other than the activation cross sections involved in the interpretation of the graphite pile experiments, PNL has not yet given detailed consideration to other cross sections involved. PNL has, however, continued to follow the progress of other obviously pertinent cross sections, e.g., Carbon, Be, postulated structural materials, and heavy elements, in their evolution as part of activities of the Cross Section Evaluation Working Group.

E. The Controlled Thermonuclear Reactor as a Neutron Source For Nuclear Transformation of Fission Products - B. R. Leonard, Jr.

The original efforts of PNL personnel in this area of transformation of fission products arose from a request to review internally a proposal⁽¹⁸⁾ to burn selected fission-products (F.P.) in a scaled-up version of the Canadian's proposed Intense Neutron Generator (ING). This review^(19,20) indicated that, aside from the feasibility of the proposed scaled-up ING, the proposal was invalid due to 1) a mis-calculation of the fission product yield of the proposed 150,000 MW(E) LMFBR economy; 2) an erroneous calculation of the neutron source available from the ING; and 3) a misunderstanding of the nuclear transmutation cross sections of the F.P. nuclei involved.

Summary

A review of alternate neutron sources led to the conclusion that at this time only a CTR device offered the potential of a sufficiently large excess of neutrons. All considerations indicated that the surplus neutrons from a CTR device represented only a marginal possibility under the assumed conditions as summarized in the references. The justification for continued consideration of the fission product transmutation concept would seem to lie in large part on the possibility that the previous assumptions will prove to be incorrect in the future economy. There are a number of possible modifications to these assumptions which might improve the feasibility of fission product transmutations. Some of these are listed in the discussion.

Discussion

A study of the feasibility of fission product burning in CTR blankets does not represent a trivial effort. The definition and optimization of a CTR blanket for fission product burning and one or more other objectives with regard to neutron economy, heat generation and removal, hazards, etc., is a problem of some magnitude. In addition, there are problems in chemical plant technology and overall systems economy to be studied. At the present time one could see a need for an effort in scope design and systems and economic studies for parametric studies of underlying assumptions.

The practicality of burning fission products in a CTR device in a blanket such as the ORNL concept seems marginal based on the following:

1. A 5000 MW (Th) device creates about 300 moles of neutrons per day.
2. Since the tritium must be regenerated the maximum excess of neutrons available for fission product burning is $300 \times (\text{Breeding Ratio} - 1)$ moles/day.
3. Postulated breeding ratio for the ORNL blanket concept are about 1.3 which gives about 90 moles/day of neutrons available for burning.
4. The postulated production of ^{90}Sr and ^{137}Cs is about 100 moles/day of each from the 1990 electric economy postulated.⁽¹⁸⁾
5. The offensive fission products are present because of their low energy (Thermal) cross sections. Consequently, it is postulated that efficiencies of utilizing the excess neutrons would be no more than some 50%.

6. Thus for DT reaction CTRs it would appear that several such plants would be required to keep abreast of the fission product production in the most optimistic case.
7. It might be anticipated that introduction of fission products into a CTR blanket would meet resistance as a potential radiological hazard.
8. Other radioactive nuclides will be built up in structural materials and other components of the CTR blanket. It must be shown that the F.P. burning is clearly greater than the buildup of other offensive radioactivity.
9. It is not yet certain that the postulated high specific neutron fluxes of CTR blankets will actually be achieved in a practical CTR because of problems of radiation damage, local power generation, etc. The postulated high fluxes are crucial to the F.P. burning concept in order to obtain nuclear transformation rates in excess of natural radioactive decay rates.

For a DD reaction CTR device where tritium production is not required the postulated F.P. burning becomes a factor of ~ 15 more feasible.

Based on the above discussion, the following points can be summarized.

1. The CTRs which operate on D, D or other reactions which do not require fuel (Tritium) breeding in the blanket clearly have an apparent neutron surplus of the required magnitude for F.P. burning.
2. A postulated economy of fission - DT fusion systems of ^{238}U or ^{232}Th burning, fissile breeding and thermal fission reactors would possibly offer a situation for the practical burning of F.P.

3. Future requirements may deem it advisable to burn F.P. in CTR blankets at a breakeven or less rate.

F. The High Temperature Lattice Test Reactor As An Experimental Facility
For Controlled Thermonuclear Reactor Blanket Studies - B. R. Leonard, Jr.

The High-Temperature Lattice Test Reactor (HTLTR)^(21,22) is a unique facility in the United States for the neutronic studies of critical fission reactor lattices at elevated temperatures (1000°C). The HTLTR was designed by Pacific Northwest Laboratories who supervised construction and operated the facility for the USAEC Division of Reactor Development and Technology. The HTLTR represents a capital cost of \$3.5 M.

The HTLTR has been used primarily for reactor physics measurements on lattices for the High Temperature Gas Cooled Reactor program and the Molten Salt Breeder Reactor program on DRDT.

Summary

The HTLTR is foreseen by PNL to be potentially a primary facility for neutronic and other technical studies of blankets for CTR devices. The HTLTR would provide a 10 ft. cube volume in which candidate blanket configurations could be assembled and maintained at controlled temperatures up to 1000°C. This volume is thermally insulated and surrounded by a radiation shield so that neutronic characteristics of blankets for D, D and D, T neutrons can be readily studied. In addition, it would provide a facility for studying the characteristics of fission blankets since it was designed as a critical fission reactor facility.

The operation of an experimental facility of this magnitude is not an inexpensive program. The use of the HTLTR as a fission reactor critical facility has required an operating budget of \$350,000/year in addition to an approximately equal amount for the scientific research and development effort. Although the operating costs are foreseen to be somewhat lower

for CTR blanket studies, the R&D program is clearly of necessity a several hundred thousand dollar/year effort.

Discussion

History of the Facility

The High Temperature Reactor Physics Program at Pacific Northwest Laboratories is a research program designed to obtain:

- Fundamental reactor neutronics data for high temperature, solid moderator, reactor lattices, and
- Nuclear engineering data in support of the economic and safe design of high temperature nuclear power reactors.

The focal point of the program centers on the use of the High Temperature Lattice Test Reactor (HTLTR) for experimental measurements. These experimental results include (a) the changes in reactor parameters caused by heating the reactor lattice of fuel, coolant, and moderator in both geometrically simple lattices as prototypical, design-point reactor cores; (b) the temperature dependence of cross sections of materials important to all thermal nuclear reactors including fuels, structural, control, and moderating materials.

The scope of the program includes checking the experimental data against the best, current theoretical models to assess the need and direction of future experimental work and theoretical development.

The HTLTR as pictured in Figure F.1 consists of a 10-ft. cube of graphite surrounded by an insulating oven. The central 5-ft. square by 10-ft. long section can be removed. As much of this region as necessary can be loaded with the test lattice of interest. The driver fuel and control systems are loaded in symmetric positions surrounding the test lattice.

Graphite heater bars extend the 10-ft. from front to back of the reactor in twenty-four locations. The oven can be sealed, evacuated, and filled with nitrogen gas prior to electrical heatup. The reactor is designed to operate at temperatures up to 1100°C.

All of the sensors, transducers, and amplifiers, essential for the process control, nuclear flux measurements, and data handling, are interfaced to the Programmed Measurement and Control System (PMACS), the heart of which is a high speed, general purpose, digital computer. PMACS includes a 19-inch three color cathode ray tube for a single point display of process variables and data storage on magnetic tape and typewritten copy. Operators manipulate and control equipment only through this system, by typing proper commands on a standard teletypewriter keyboard which are accepted by the computer programs and executed automatically. Closed loop control is provided for the heating system and the gas system. The nuclear system is also computer controlled except that the operator must manually operate a switch to move control rods that he had previously selected by the typewriter.

Three major experiments have been completed in HTLTR in support of the High Temperature Gas-Cooled Reactor Program. Test lattices fueled with ThO_2 - $^{235}\text{UO}_2$ and ThO_2 - $^{233}\text{UO}_2$, and a MSBR lattice were studied, over the range from room temperature to 1000°C. The results derived from the measurements include the variation of k_{∞} as a function of the temperature of the test assembly, and the Doppler coefficient for the thorium in the test lattices. Additional experiments in support of the HTGR program include a lattice fueled with ThO_2 - PuO_2 .

FIGURE F.1 High Temperature Lattice Test Reactor

Description and Design Specifications (HTLTR)

The reactor was designed to study the lattice properties of both gas-cooled and liquid-cooled fuel elements in a solid crystalline moderator. The coolant used in the experimental core is nitrogen. The concept uses a solid graphite moderator. The fuel elements used included only those suitable for high temperature and long exposures.

The building facilities consist of:

1. A reactor room with basement and an attached enclosure for the neutron beam and analyzer.
2. A service building attached to the reactor with basement, main floor and second floor.
3. An exhaust stack (40' high).
4. A liquid nitrogen storage facility (8,000 gal capacity).
5. Cooling tower.

The moderator block is a 10-foot cube of graphite enclosed within a thermally-insulated, gas-tight container. The reactor was designed to operate initially at 1000°C and to be capable of operation at 1200°C with some additional equipment. The insulation and the gas-tight vessel is equipped with sealed doors that can be removed for access to the reactor.

The reactor is heated electrically. Three-hundred-eighty-four kW of electrical power are delivered to graphite heating elements. Eight saturable reactors regulate the electrical power input. Graphite heating elements are distributed to provide uniform heat.

The experimental equipment within and attached to the reactor permits the removal of the 10' long parallelepiped experimental core section whose maximum cross section is 5' x 5'. This experimental core contains a test

cell (about 15" x 15" x 78") which can be moved along its center line.

In addition, a small oscillator is attached to the front face to oscillate samples up to five pounds in weight while the reactor is at temperature.

The reactor gas system is a closed nitrogen circulating loop containing equipment to circulate gas for temperature leveling and shutdown cooling. The gas system also contains instrumentation for detection of gases, transient radioactivity, and water leaking from the primary heat exchanger. The design life of all reactor process components is based on a life span of about ten years.

The design includes adequate provision to attain, measure and record the following operating temperatures:

Over-all reactor	1000 ⁰ C
Central test fuel element	1500 ⁰ C
	(temperature measurement and indication only)
Heater surfaces	1600 ⁰ C maximum

Electrical heating is adequate to heat the entire moderator and thermal shielding to equilibrium, with a temperature of 1000⁰C of the moderator, in five days (120 hours). Heat is removed by circulating nitrogen gas, through water cooled heat exchangers. A containment enclosure surrounds the reactor and limits maximum leakage to .5% of volume per hour at 1 psig. The detection of radioactive materials in the system is accomplished by continuous scanning of the purge system and reactor gas circulating system. The graphite material used for this project is the type SGBF and TSGBF. The bars are 4-3/16 inches squares by about 4 feet long.

A neutron chopper is available for energy distribution measurements.

A beam of neutrons from a surface on the test lattice is allowed to pass through a hole in the side of the reactor. Just outside the reactor the beam encounters a spinning rotor. The rotor is pierced perpendicularly to its spinning axis by a set of parallel slits which permit the transmission of short bursts of neutrons down a flight path. The neutrons that pass through the rotor traverse a flight path of known length and impinge on a detector. The time of flight of a neutron is a measure of its velocity and the number of arrivals in a short time interval is recorded in a series of time channels. A neutron distribution in velocity is thus obtained from which the neutron energy spectrum can be derived.

Facility functions are as follows:

1. Measures at any temperature up to 1000°C the neutron spectrum emerging from any selected region of an experimental lattice.
2. Provides the primary method of determining when the spectrum of neutrons in the experimental lattice has been adjusted to what it would be in an infinite lattice.
3. One of the important nuclear properties of a lattice, which is basic to understanding it, is the spectrum of the neutrons at various positions in the lattice. It is not accurately calculable and hence measurement of it is mandatory, particularly at elevated temperatures, or over large temperature ranges.

In addition to a measurement of the neutron spectrum in prototype test lattices, which are too complicated to calculate, fundamental problems in neutron slowing down theory can also be studied with simpler test cell arrangements.

The bulk of information on neutron spectra is for one moderator temperature. In HTLTR one has a means of studying spectra over

FIGURE F.2 The Control Room for the HTLTR. The Controller Readout Panels are, from Left to Right: Radiation Monitoring, gas analysis, the Van de Graaff Neutron Generator, the Safety Circuit, the Operator's Control Typewriter, and the three-Color Alphanumeric CRT Display of Process Variables, the Auxiliary Safety Channels, the PDR-7 Computer with Console, and the Output on Logging Typewriter.

FIGURE F.3 The High Temperature Lattice Test Reactor. A View of the Rear Face, Showing 9 Control Rod Drives, the Heavy-Duty Oscillator, and a Five-Foot Access Door to the Reactor Core. On the Top of Reactor are Four Vertical Safety Rods and the Neutron-Sensing Ionization and BF_3 Chambers.

a wide range of temperatures in ranges where the greatest uncertainty exists. With this additional dimension it is hoped that a better understanding of lattice spectra can be obtained.

Facility Modifications

The HTLTR will be modified in order to be more efficiently utilized in the study of the neutronics characteristics of Controlled Thermonuclear Reactor (CTR) blanket designs. Expected CTR power plants will have blanket regions surrounding the hot plasma. First generation plasmas will give off the bulk of their energy in the form of 14 MeV neutrons. The blanket services as a moderating media, a heat transfer media, and in addition, a region for breeding of makeup fuel for the device. The current knowledge of neutronics characteristics of 14 MeV neutrons in hot (1000°C) blankets for the materials of interest is insufficient to accurately design the blankets. Accurate blanket design is crucial because:

- Blanket thicknesses will determine the size of the surrounding superconducting magnets. The superconducting magnet is the over-riding capital cost item of the reactor.
- Blanket designs will determine the breeding ratio of the device. This not only has economic value but also determines fuel inventory which is in turn a strong consideration in the safety analysis.
- The dynamic characteristics of the blanket will be determined by the design.

The HTLTR is a unique facility for making such measurements. It is capable of being heated to cover the whole range of expected CTR blanket temperatures. Materials of interest to the program can be readily introduced into the facility to mockup appropriate blanket designs. A low level source of 14 MeV neutrons is available. The facility will be used as a

neutron scattering media and will thus have the fissionable fuel removed during these experiments. The program has begun with the initiation of benchmark experiments and will proceed to more complicated design confirmation experiments.

It is proposed to modify the HTLTR facility for the experiments in the following manner:

- Design, procure, and install a data collection system in the facility for measuring the neutron flux as a function of energy and position.
- Design and install a port for placing the source in the center of the facility. Current work is being done with the source at the edge of the facility.
- Procure and install a more intense 14 MeV neutron source.

By this approach, a flexible and unique facility for obtaining CTR blanket design data can be developed at minimum cost to the USAEC CTR program, utilizing existing facilities. A comparable facility, of crucial importance to the program, would require a much larger expenditure of funds.

G. Neutron Damage to Superconductors - D. L. Lesser

Neutron damage to superconductor materials with subsequent superconducting property changes is one of the most significant CTR engineering problems. Experiments reported in the literature using reactor neutrons give an indication of the nature of neutron damage to superconductors and neutron fluences and energies at which the effects are likely to occur. Previous work indicates that changes in superconductor properties in niobium^(23,24,25) appear with a fluence of approximately 10^{14} fast neutrons/cm², with a saturation effect in the changes at a fluence of 10^{17} - 10^{19} fast neutrons/cm². Only neutrons with energies in excess of 1 MeV are believed to contribute. Wolkenhauer⁽²⁶⁾ anticipates an uncollided flux of 2.0×10^{12} n/cm² sec at the magnets in typical fusion reactor geometries, assuming a flux of 4×10^{14} n/cm² incident on the vacuum wall. Hence, extensive changes to superconductor properties could be expected to occur in a few minutes to a few days. The changes are (1) an increase in the upper critical field, (2) an decrease in the transition temperature, and (3) a greater irreversibility of magnetization curves from flux pinning on defects. It would appear desirable to perform experiments on candidate superconducting magnet materials for fusion reactors in fluences up to those expected on a fusion reactor superconducting magnet. Marginal fast neutron flux from (charged particle, neutron) interaction with available accelerators led to a decision to defer planning of an experiment.

A study by O.K. Harling⁽²⁷⁾ indicates the feasibility of cryogenic temperature irradiation in a fast flux of 10^{14} neutrons/cm²/sec at a location two to three feet above the core of the Fast Test Reactor. The accompanying nuclear heating of .5 watt/gm does not impose prohibitive cooling requirements. This facility would be excellent for the proposed studies.

H. Fusion Cross Sections - B. H. Duane

This work has been focused on development of a controlled thermonuclear reactor (CTR) nuclear data file upon which to base CTR analysis studies. The work involves nonlinear least squares fitting of relevant cross section measurements to quantum theory functional forms describing charge barrier penetration followed by nuclear resonance interaction.

Theory

Previous work of this sort, as reviewed by Post⁽²⁸⁾ and summarized by Glasstone⁽²⁹⁾, appears to have been limited to use of Gamow's⁽³⁰⁾ charge barrier penetration probability

$$p = (q_1 q_2 / 2\epsilon_0 \hbar v) \exp(-q_1 q_2 / 2\epsilon_0 \hbar v), \quad (H.1)$$

which is merely the low energy limit of Mott's⁽³¹⁾ more broadly relevant charge barrier penetration probability

$$p = (q_1 q_2 / 2\epsilon_0 \hbar v) / [\exp(q_1 q_2 / 2\epsilon_0 \hbar v) - 1]. \quad (H.2)$$

In both expressions, v is the incident approach velocity of charges q_1 and q_2 , $\hbar$ is the angular momentum quantum, and ϵ_0 is the dielectric permittivity of space (in rationalized mks units). (In cgs units, ϵ_0 is replaced by $1/4\pi$.) Mott derives the charge barrier penetration probability (H.2) by use of Temple's⁽³²⁾ exact solution of the Schroedinger wave equation for two charged particles (as a confluent hypergeometric wave function of parabolic coordinates).

Observing that the high energy limit ($v = \infty$) of Gamow's penetration probability (H.1) is zero (as it manifestly should not be), and that the high energy limit of Mott's penetration probability (H.2) is unity (as it

manifestly should be), may help to explain persistent previous failures of Gamow fits to follow fusion cross section measurements satisfactorily above 13-20 KeV.

Application

Multiplying Mott's charge barrier penetration probability (H.2) by a flexibly truncated summation of Breit-Wigner⁽³³⁾ nuclear resonance cross sections leads to fitting fusion cross section measurements $\sigma(E)$, as a function of laboratory bombardment energy E , to the theoretical functional

$$\sigma(E, \underline{A}) = [E \{ \exp(A_1/\sqrt{E}) - 1 \}]^{-1} \sum_{n=1}^N A_{3n-1} / [1 + (A_{3n}E - A_{3n+1})^2]. \quad (H.3)$$

The parameters $\underline{A} = (A_1, A_2, A_3, A_4, A_5, \dots)$ appearing here are evaluated by nonlinear least squares fitting of this functional form to available measurements. As computer programmed in BNW Program Learn-III, the parameter variation proceeds by iterative minimization of an inverse-variance-weighted sum-of-squares of discrepancies between measurements $\sigma_m(E_m)$ and theory $\sigma(E_m, \underline{A})$, of the form

$$Q(\underline{A}) = \sum_{m=1}^M [\sigma_m(E_m) - \sigma(E_m, \underline{A})]^2 / [\delta\sigma_m(E_m)]^2, \quad (H.3)$$

where $\delta\sigma_m(E_m)$ is the root-mean-square measurement uncertainty for $\sigma_m(E_m)$.

Preliminary results for two of the more important fusion reactions are presented in both graphical and tabular form in Figs. H.1 and H.2. Both the 5-parameter fit to the $T(d,n)He^4$ reaction and the 4-parameter fit to the $D(d,p)T$ reaction were found to lie at points of sharply diminishing returns in analysis refinement, in the regression theory sense that fewer parameters drastically deteriorate the quality of the fit. For the $T(d,n)He^4$ reaction,

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a 7-parameter pass did not improve the outlier discrepancy prominent at 10.5 MeV.

Fig. H.1: $T(d,n)He^4$ FUSION CROSS SECTION σ AS FUNCTION OF ENERGY E

Measurements: ϕ : LA-2014 Mar 1956

Theory: $\equiv$: Nonlinear least squares fit, BNW Prog. LEARN (BNWL-390)

$$\sigma = [E \{ \exp (A_1/\sqrt{E}) - 1 \}]^{-1} [A_2 / \{ 1 + (A_3 E - A_4)^2 \} + A_5]$$

$$A_1 = 1453 \pm 14 \quad \sqrt{eV}$$

$$A_2 = (5.02 \pm .26) \times 10^7 \text{ eV} \cdot \text{barn}$$

$$A_3 = (13.67 \pm .45) \times 10^{-6} \text{ /eV}$$

$$A_4 = 1.076 \pm .055$$

$$A_5 = (4.08 \pm .15) \times 10^5 \text{ eV} \cdot \text{barn}$$

Fig. H.2. D(d,p)T FUSION CROSS SECTION σ AS FUNCTION OF ENERGY E

Measurements: ϕ : LA-2014 Mar 1956

Theory: $\equiv$: Nonlinear least squares fit, BNW Prog. LEARN (BNWL-390)

$$\sigma = [E \{ \exp(A_1/\sqrt{E}) - 1 \}]^{-1} [A_2 / \{ 1 + (A_3 E - A_4)^2 \}]$$

$$A_1 = 1457.7 \pm 5.0 \quad \sqrt{\text{eV}}$$

$$A_2 = (372 \pm 10) \times 10^3 \quad \text{eV} \cdot \text{barn}$$

$$A_3 = (4.36 \pm .26) \times 10^{-7} \quad / \text{eV}$$

$$A_4 = 1.220 \pm .038$$

I. Reaction of Hydrogen and Deuterium with Niobium - Zirconium Alloys at 900°C - L. A. Charlot and R. E. Westerman

As a result of an investigation of sputtering rates of niobium alloys by D^+ ions, Yonts⁽³⁴⁾ reported that a Nb-1 Zr alloy showed a comparatively strong tendency to capture and retain deuterium at high temperatures. The kinetics of absorption and permeation of niobium-zirconium alloys by hydrogen isotopes are extremely important to D - T thermonuclear reactor design. For this reason a cursory study was undertaken to determine the extent to which zirconium alloy additions might affect the normally rapid rates of diffusion of hydrogen and deuterium through niobium at a temperature near that anticipated for the vacuum wall of an operating thermonuclear reactor.

Summary

The present study does not indicate any unusually high temperature absorption or desorption kinetics of hydrogen or deuterium by niobium alloys caused by the presence of zirconium. It was felt that the experimental techniques employed in the present study were not able to discriminate between alloys, or, for that matter, between hydrogen or deuterium, hence the data boundaries have been used exclusively instead of data points. It is certain that differences in reaction kinetics will be caused by the presence of alloy elements such as zirconium and yttrium; however, it remains for a more detailed, extensive study to identify and quantify these differences.

Discussion

The experimental approach involved determining the kinetics of absorption of H_2 or D_2 gas by an alloy sample of prescribed geometry, then outgassing the sample and determining the kinetics of desorption. The absorp-

tion kinetics were determined by monitoring a pressure decay; a pressure buildup was monitored in the case of desorption. In the latter case a mercury diffusion pump extracted the gas from the sample and held it in a standard volume for measurement.

The absorption studies led to information concerning pressure-composition equilibria; this data is also included.

Four alloy buttons were made up by standard melting procedures, and cylinders 0.64 cm x 3.18 cm long were machined from the buttons. The samples were vapor blasted to minimize the possibility of a slow surface reaction and degreased prior to test. The samples had the following nominal compositions:

- Nb (commercially pure)
- Nb-10w/oZr
- Nb-10w/oZr-0.1w/oY
- Nb-20w/oZr

Yttrium was included because it is an expected niobium transmutation product; zirconium can result from transmutation, or it may be intentionally added as an alloying element.

The samples were suspended in a quartz reaction tube, and outgassed at $900 \pm 5^\circ\text{C}$. A known quantity of H_2 (commercial purity) or D_2 (from a heated UD_3 bed) was then admitted into the chamber. The reaction rate was determined by monitoring the pressure decay on an oil manometer.

The quantity of gas initially admitted was that amount expected to result in a final concentration in the sample of ≈ 100 ppm by weight, taking into account the final equilibrium pressure. This final pressure is not well known in the case of Nb-Zr alloys, and so the 100 ppm level

was not attained in all cases. Normalization of the kinetic curves made the absolute level attained relatively unimportant.

Albrecht, et.al⁽³⁵⁾ developed an analytical expression relating fractional uptake of gas to a function of diffusivity D , time t , and cylinder radius a ; they found that a plot of C/C_0 vs. $(Dt/a^2)^{1/2}$ was linear up to about 40% saturation. (In practice, the approximation to linearity is fairly good even to 70%.) Such a plot then leads to an easy determination of diffusivity. Albrecht, et.al., used an expression specifically developed for a finite specimen having a length/radius (l/a) ratio of 5. Crank⁽³⁶⁾ presents a plot of the same relationship for a cylindrical sample having an l/a ratio of infinity; the two plots are indistinguishable in the region of interest indicating the applicability of the overall approach to the present study where the sample l/a ratio is 10. It is important to note, however, that the diffusivities obtained by this method in the present study must be considered approximate values only, as the analysis discussed in the foregoing is not strictly applicable when a steady diminution in the surface concentration (C_0 by definition also the final sample concentration) takes place during the absorption reaction. This consideration is not important in the case of niobium or lightly alloyed niobium, as the initial and final gas pressures, hence initial and final surface solute concentrations, are quite close. In the case of a Nb-20Zr alloy, however, the initial and final pressures are markedly different, so that the surface solute concentration can vary substantially during the experiment. This will lead to an artificially high apparent diffusivity for the Nb-20Zr alloy, estimated to be of the order of a factor of two in the present study. It was felt that this inherent error was acceptable

within the modest scope of the present effort. Also, the pressure decay method is especially well suited to the study of rapid absorption rates, such as those encountered in the present study, when the reaction was commonly 40% complete within 30 seconds.

After a given absorption study had been completed, the sample was quickly withdrawn from the reaction chamber and allowed to cool. During cooling more gas was absorbed; this was measured and "added" to the sample's inventory for the desorption study. The sample was then dropped back into the furnace tube held at $900 \pm 5^{\circ}\text{C}$ and a mercury diffusion pump used to transfer the gas from the sample into a standard volume for measurement. Outgassing and gas collection began before the sample reached 900°C . The outgassing was generally continued until more than 99% of the gas inventoried in the sample was recovered.

The concentration of hydrogen and deuterium in the alloys in equilibrium with the final pressures of H_2 or D_2 at $900 \pm 5^{\circ}\text{C}$ is shown in Table I.1. Concentrations are given in weight units.

As expected, solubilities of deuterium are slightly less than hydrogen on an atomic basis. The strong affinity of alloy element zirconium for hydrogen and deuterium relative to niobium is evident.

TABLE I.1
PRESSURE-COMPOSITION EQUILIBRIA

<u>Alloy</u>	<u>Final P_{H₂} Torr</u>	<u>Conc. H (C_O) ppm by wt⁰</u>	<u>Final P_{D₂} Torr</u>	<u>Conc. D (C_O) ppm by wt⁰</u>
Nb	67.36	73	64.02	123
			63.83	123
Nb-10Zr	5.58	54	5.55	102
			5.56	100
Nb-10Zr-0.1Y	1.25	24	1.30	45
			1.48	43
Nb-20Zr	2.15	111	2.28	200

The gas absorption kinetics are shown graphically in Figure I.1. All absorption curves fit within the boundaries shown. Duplicate trials on a given specimen showed good reproducibility. The diffusivity values calculated for hydrogen fall within the boundaries $D = 1.6 \times 10^{-4} \text{ cm}^2/\text{sec}$ and $D = 2.7 \times 10^{-4} \text{ cm}^2/\text{sec}$; the values for deuterium fall within the boundaries $D = 1.1 \times 10^{-4} \text{ cm}^2/\text{sec}$ and $D = 2.6 \times 10^{-4} \text{ cm}^2/\text{sec}$. An extrapolation to 900°C of the data of Albrecht, et. al.⁽³⁵⁾ for hydrogen diffusion in niobium yields a value $D = 3.9 \times 10^{-4} \text{ cm}^2/\text{sec}$.

Desorption kinetics as determined in the present study are not readily amenable to mathematical analysis because the tests are non-isothermal and because the solute gradient in the sample is not well defined prior to gas extraction. However, inter-sample comparison may be expected to yield data on the relative ease of recovery of gas from the individual alloys. The boundaries shown in Figure I.2 contained all of the data obtained for desorption of deuterium and hydrogen from all of the alloys.

The curves were normalized such that $t = 0$ corresponds with the first appearance of outgassing, which generally began within two minutes of the sample introduction into the furnace tube.

The data band of Figure I.2 indicates that the outgassing kinetics of all systems studied follow the same general pattern, with no unusual gas retention exhibited by any given alloy.

FIGURE I.1 Absorption of Hydrogen and Deuterium by All Alloys at $900 \pm 5^\circ\text{C}$

FIGURE I.2. Hydrogen/Deuterium Desorption Kinetics,
All Alloys. Outgassing First Sensed at $t=0$.

J. Irradiation Induced Swelling of Controlled Thermonuclear Reactor Materials - G. L. Kulcinski and J. L. Brimhall

The materials used to construct the first Controlled Thermonuclear Fusion Reactors (CTR) will be subjected to the most severe high temperature radiation damage conditions that have even been imposed on structural components.^(37,38) For example, the inner wall of the plasma containment vessel will be subjected to 14 MeV neutron currents of 4×10^{13} to 4×10^{14} n/cm²/sec and total neutron fluxes which are an order of magnitude higher. Copious amounts of high energy helium, deuterium and tritium atoms will also bombard the wall surface at the same time that it is under intense photon irradiation. Such irradiations will cause each atom to be displaced from its lattice site several hundred times per year. The high (n,p) and (n, α) cross sections for 14 MeV neutrons will cause several hundred atomic parts per million of helium and as much as 1000-2000 atomic parts per million of hydrogen gas to be generated within the samples.

Such a high rate of defect and impurity atom generation at the anticipated operating conditions (600-1000°C) of the CTR will result in the precipitation of the vacancies and gas atoms into voids and gas bubbles respectively. These latter defects will have two main detrimental effects on metals; they will induce significant dimensional changes which will introduce high stress levels, as well as stress gradients in metals, and they will cause structural metals to become extremely brittle, unable to withstand even a few percent of strain before failure. Present work has provided information on the dimensional stability problem and future studies are planned to investigate the embrittlement aspects.

Preliminary analysis of the swelling problem quickly revealed that, even if the corresponding CTR temperatures could be reproduced in present day fission reactor facilities, it would take roughly 3 - 4 years of fission reactor exposure to produce the displacement damage found after only one year of CTR wall exposure (for a 10 MW/m^2 wall loading). Furthermore, fission reactor irradiations will not be able to simulate the high gas production rates characteristic of 14 MeV neutrons.

Scientists have therefore turned to an alternate method of simulating high damage states, namely the use of external beams of high energy heavy ions.^(39,40,41) This technique allows the investigator to increase the damage rate within the solid by 3 to 4 orders of magnitude over that characteristic of present day fission reactors. Such high rates of damage allow one to form in a matter of hours, high temperature damage states which would normally take years of fission or fusion reactor irradiation to produce. There are several articles⁽⁴²⁻⁴⁹⁾ in the literature describing the use and limitations of this technique. Most of the previous studies⁽³⁹⁻⁴⁹⁾ have been aimed at simulating high fluence damage anticipated in future LMFBR's, but it is obvious that these techniques can also be used for CTR materials as well.

Personnel at Pacific Northwest Laboratories have conducted such simulation studies on the leading candidates for fission reactor structural materials;^(45,50,51) pure Nb, Mo, V and selected alloys of these elements. The preliminary results of these experiments are summarized here. In particular, the effect of irradiation temperature on the production of voids in Nb and Nb-Zr alloys, Mo and TZM (a titanium, zirconium modified Mo alloy) and V in the absence of helium and significant hydrogen gas atoms has been investigated. Future studies will include the simultaneous injection of these

same metals with He or H atoms while the samples are being bombarded with high energy heavy ions.

Summary

Transmission electron microscopy investigations of irradiated fusion reactor materials has revealed that swelling values of up to 1% will be produced in less than 1 month of exposure to future CTR radiation environments. Such swelling, and that induced by longer term irradiation could cause significant dimensional changes and increases in the absolute stress levels as well as stress gradients in the thin wall containment vessels proposed in current CTR designs. It is also shown that contrary to previous proposals, inert gas atoms, such as helium, are not essential in the nucleation of voids. However, the inclusion of large amounts of neutronically produced helium and hydrogen in metals may increase the severity of the swelling problem and should be investigated.

Experimental Procedure

The chemical compositions of the materials used for this study are given in Table J.1. Niobium alloys of nominally 1, 5 and 10 atomic percent were also prepared from the starting Nb stock. The typical samples used for this study were 3 mm in diameter and varied in thickness from 0.07 mm to 0.25 mm. Pre-irradiation anneals for 1/2 hour at 1800°C were given to all the samples except for the vanadium which was annealed at 1100°C. The vacuum in all cases were varied from 1 - 2×10^{-6} Torr.

The experimental details of the bombardment techniques are described elsewhere.⁽⁴⁵⁾ Briefly, the 3 mm diameter samples were irradiated with 7.5 MeV Ta ions or 5 MeV Ni ions while the samples were heated to 650 - 1000°C on a tantalum furnace. In the case of the Ni ion bombardment, the ion fluence was held constant at $7 \times 10^{15} \text{ cm}^{-2}$ in order to produce a con-

TABLE J.1

PRE-IRRADIATION PROPERTIES
OF MATERIALS FOR ION BOMBARDMENT STUDIES

<u>Impurity</u>	<u>Mo</u>	<u>PPM (wt) TZM</u>	<u>Nb</u>	<u>V</u>
Al	<5	<300	100	20
C	200	100-300	30	250
Cd	<10			
Co	<5			
Cr	<5		60	100
Cu	<2			6
Fe	100	<300	60	60
H	0.8	1-10	2	10
Mg	<1	<100	0.8	
Mn	<2			
Mo	Bal.	Bal.		
N	1	4-15	90	290
Ni	<5		9	
O	4.5	10-20	145	570
Pb	<10			
Si	<10	<300	20	100
Sn	<5			6
Ta			200	10
Ti	<10	4200-4700	60	3
V	<5		5	
W	150		80	10
Zr	<20	800-1100	60	

stant damage state of approximately 5 dpa (displacements per atom, i.e., the number of times each atom is displaced from its lattice site during irradiation). The average beam current was 1.2×10^{13} ions $\text{cm}^{-2} \text{sec}^{-1}$ which resulted in a displacement rate of approximately $8 \times 10^{-3} \text{sec}^{-1}$. The total Ta ion fluence was also 7×10^{15} ions cm^{-2} but the beam current in this case was 2.5×10^{12} ions $\text{cm}^{-2} \text{sec}^{-1}$ which produced damage at approximately $1.2 \times 10^{-2} \text{sec}^{-1}$.

The amount of displacement damage was calculated^(44,52) as a function of penetration into each of the metals and Figure J.1 represents a typical calibration curve used for Mo and TZM in this study. The curves for Nb and V are only slightly different and not repeated here. The techniques for removing a fixed amount of the damaged surface are also described elsewhere,^(46,53) and the resulting microstructure was observed with a model Hitachi 200 F microscope. The average void size was determined from 160,000x photomicrographs and the number density of voids was determined by stereo microscopy. The resultant volume change was calculated from the experimental data.

Results

The results of the microstructural examinations after the Ni ion bombardment to approximately 5 dpa are listed in Table J.2 and those for the Ta ion bombardment to approximately 20-100 dpa are shown in Table J.3. It is convenient to describe the results obtained at a relatively constant damage level first.

TABLE J.2

VOID DATA ON NICKEL BOMBARDED REFRACTORY METALS^(a)

<u>Irradiation Temp., °C</u>	<u>Average Void Size A</u>	<u>Average Void Density $\times 10^{16}/\text{cc}$</u>	<u>Calculated Swelling %</u>
<u>Molybdenum</u>			
650 ^(b)	ND ^(c)	ND	--
800	50	5.2	0.34
900	75	4.7	1.0
1000	120	0.53	0.48
<u>TZM</u>			
800	60	3.2	0.36
900	80	1.6	0.43
1000	105	0.36	0.22
<u>Niobium</u>			
650 ^(b)	ND	ND	--
700	ND	ND	--
800	45	6	0.29
900	220	0.15	0.83
1000	350	0.018	0.34

(a) Damage level approximately 5 dpa produced by 5 MeV Ni ions except where indicated.

(b) 6 MeV Ni ions.

(c) ND - None detected.

TABLE J.3

PRELIMINARY VOID DATA ON
TANTALUM BOMBARDED REFRACTORY METALS

<u>Irradiation Temp., °C</u>	<u>Average Void Size A</u>	<u>Average Void Density x 10¹⁶/cc</u>	<u>Calculated Swelling %</u>	<u>dpa</u>
<u>Vanadium</u>				
700	440 ^(a)	0.027	2.3	30
800	665 ^(a)	0.01	2.9	22
900	ND	ND	--	22
900	ND	ND	--	83
<u>Niobium</u>				
700	ND	ND	--	25
800	Analysis currently underway.			31
900				28
900				76
<u>Nb-1 Zr</u>				
700	40	3.8	0.13	34
800	65	1.9	0.2	30
900	220	0.2	1.12	26
900	285	0.2	2.45	100
<u>Nb-5 Zr</u>				
700	Analysis currently underway			32
800				26
900				34
900				78
<u>Nb-10 Zr</u>				
700	Analysis currently underway.			~30
800				~30
900				~30
900				~80

(a) Cube edge.

Voids were found in pure Mo and Nb, and TZM after approximately 5 dpa irradiations at 800, 900 and 1000°C. Figure J.2 shows typical void structures in Nb and Mo after the 900 and 1000°C irradiations and Figure J.3 shows the voids produced in TZM over the 800 - 1000°C range. No voids, only dislocation loops, were found in Nb and Mo irradiated at 650 and 700°C.

The average void size in the metals increases with increasing irradiation temperature while the number density decreases. The resultant volume change for the metals goes through a maximum at approximately 1% at 900°C (Figure J.4). Preliminary error analysis of these values indicate that error bars of $\pm 50\%$ because of the large errors encountered in measuring foil thickness. On this basis, we would say that there is little difference in the swelling of pure Mo and Nb over this temperature range and there is possibly a slight reduction of swelling in TZM after 900°C irradiation.

The morphology of the voids in the Mo and TZM samples is difficult to determine because of the small void size (50 - 120 Å). However, the voids in Nb grow to a sufficient size during irradiation at 1000°C to determine that they are cubes bounded by {100} planes. Figure J.5 shows the voids in Nb when the foil plane is near a {110} and a {111} orientations (see illustrations in Figure J.5).

There is also evidence for a nonrandom configuration of voids in the 800°C irradiated Nb samples. Figure J.6 shows the voids in a {110} foil orientation. The voids appear black in this picture because of absorption effects. Note the alignment of voids perpendicular to the [101] direction. (This can be seen more easily if the figure is viewed endwise). This arrangement and that determined for {100} orientations indicates that the

voids are in a bcc superlattice that is coincident with the crystal lattice and has a lattice constant of approximately 350°A .

Irradiation of V and Nb-Zr alloys with 7.5 MeV Ta ions also produced voids at temperatures of interest to CTR designers (Figures J.7 & J.8). The voids in V at 700 and 800°C were quite large and tended to be $\{100\}$ cubes. The cube edges were 440 and 665°A for the 700 and 800°C irradiations, respectively, which resulted in swelling values between 2 - 3% (Table J.3). No voids were found after a 900°C irradiation to 20 or 80 dpa. A ribbon like precipitate was found in the 700°C bombarded sample (Figure J.7). The long dimension of the precipitate appears to be parallel to the $\{100\}$ directions.

Investigation of the pure niobium samples bombarded to 30-75 dpa had not been completed at the time this report was written, but it has been established that no voids are found in the Nb irradiated to 30 dpa at 700°C , similar to what was found after a 5 dpa nickel ion bombardment at 700°C (see Table J.2).

In contrast to pure niobium, voids were produced in a Nb-1 Zr alloy during a 700°C irradiation to approximately 30 dpa (see Figure J.8). The voids were quite small (approximately 40°A diameter) and present in a rather large density ($3.8 \times 10^{16} \text{ cm}^{-3}$). Irradiation at 800°C resulted in an increased void size (65°A) and a reduction in void density to $1.9 \times 10^{16} \text{ cm}^{-3}$. The average void diameter increased to 220°A during a 900°C - 26 dpa irradiation and the number density of voids was further reduced to $0.2 \times 10^{16} \text{ cm}^{-3}$. The total volume change increased with irradiation temperature up to 900°C , similar to the trend found during lower fluence data on pure Nb (see Figure J.8 and Table J.2). However, the

total swelling in the Nb-1 Zr alloy at 900°C was only 35% higher than that found for pure Nb at the same temperature even though the total damage was approximately 500% higher.

Studies are currently underway to evaluate the Nb-5 Zr and Nb-10 Zr alloys.

Discussion

The results of this study show that voids may be easily produced in refractory metals by heavy ion bombardment at typical CTR operating temperature. It is also important to note that these voids can be formed without the aid of any inert gas atoms such as helium. The exact mechanism for nucleation of voids in metals is still not known, but recent experiments by Norris^(54,55) and Urban⁽⁵⁶⁾ have eliminated the concept of homogeneous nucleation by displacement spikes or by the agglomeration of vacancies in a supersaturated lattice. One of the most probable remaining mechanisms is the nucleation of voids by impurity gas atoms but not necessarily by noble gases. The present results do not prove or disprove that theory, but there is certainly sufficient residual (O + H + H) gas atoms in all three metals for more than one hundred atoms per void in the 5 dpa studies. Future work on specially degassed samples will aid in testing this concept.

In order to extend to present work to the higher damage levels, it is certainly necessary to establish that the microstructures already produced in a few minutes to a few hours of ion bombardment are comparable to the long term neutron irradiation data reported in the literature. This is best accomplished by a comparison of the void induced swelling in terms of irradiation temperature and fluence. Before the comparison can be made, it is important for the reader to recognize that the high defect production rates in this type of study (10^{-3} to 10^{-2} sec⁻¹) compared to those actually encountered in CTR reactors (10^{-6} to 10^{-5} sec⁻¹) will result in an

"effective" irradiation temperature somewhat lower than the actual furnace temperature. Bullough and Perrin⁽⁵⁷⁾ have shown that in order to compare the present results obtained at some ion bombardment temperature, T_i , to those which would be obtained by neutron irradiation at temperature of T_n , one must make the following correction.

$$T_n = \frac{T_i}{1 + \frac{T_i k}{E_v} \ln \frac{K_i}{K_n}} \quad (J.1)$$

where

E_v is the self diffusion energy of vacancies

k is Boltzmann's constant

K_i is the defect production rate during ion bombardment

K_n is the defect production rate during neutron irradiation.

If one uses the self diffusion energies of 4.0 eV⁽⁵⁸⁾ for Mo, 4.12 eV⁽⁵⁹⁾ for Nb and 3.12 eV⁽⁶⁰⁾ for V, and $K_n \sim 3 \times 10^{-6} \text{ sec}^{-1}$ for CTR reactors⁽⁴⁸⁾ then the present ion bombardment temperatures correspond to the effective temperatures listed in Table J.4. The threshold for void formation in terms of effective temperatures is then approximately 600°C, close to what Brimhall and Mastel find for the same stock material.⁽⁵¹⁾ The lack of void induced swelling at 650°C in the ion bombarded samples can then be explained on the basis that no voids have yet been discovered after neutron irradiation at 520°C. The magnitude of the swelling in this study is comparable to the neutron irradiated material if it is normalized on the basis of equivalent dpa values. Brimhall, et.al.,⁽⁶²⁾ find the volume change ranges from 0.07 to 0.08% over the 650 - 750°C temperature

TABLE J.4

EFFECTIVE IRRADIATION TEMPERATURE
FOR FUTURE CTR REACTORS IN TERMS OF
PRESENT ION BOMBARDMENT STUDIES

Ion Bombardment Temperature	"Effective" CTR Temperature, T _n °C ^(a)		
	Mo	Nb	V
650	522 (0.28) (b)	525 (0.30)	492 (0.35)
700	558 (0.29)	562 (0.31)	526 (0.37)
800	630 (0.31)	635 (0.34)	592 (0.40)
900	700 (0.34)	705 (0.36)	656 (0.43)
1000	768 (0.36)	774 (0.39)	717 (0.46)

(a) For K_n approximately $3 \times 10^{-6} \text{ sec}^{-1}$

(b) Fraction of absolute melting point.

range for a 1.5×10^{21} n/cm² (E>0.1 MeV) irradiation. This neutron fluence would amount to a dpa value of approximately 1.5 if a Kinchen and Pease⁽⁶³⁾ model is valid. If the swelling increase in Mo follows roughly the 3/2 dependence on fluence,⁽⁴⁶⁾ then the neutron irradiated samples would swell 0.4 - 0.5% at 5 dpa in the 650 - 750°C range. This swelling is reasonably close to the 0.4 - 1.0% swelling over the 630 - 770°C "effective" temperature range for the present experiment.

The present results also agree fairly well with previously reported data for neutron irradiated TZM. Brimhall, et.al.,⁽⁶²⁾ have reported that voids form in neutron irradiated TZM at 615°C, but that only precipitates, not voids, were formed during 750 and 900°C studies. We found both precipitates and voids in ion bombarded TZM in this temperature range and speculate that the time of irradiation may account for the differences. It would be expected that if the precipitates form by a long term aging process, in which they absorb the vacancies produced by neutron irradiation, then the higher super-saturation present during ion bombardment might provide enough vacancies for both the precipitates and voids to form.

There is comparatively little high temperature neutron irradiation data on Nb and Nb-Zr alloys to compare with the present results. Wiffen⁽⁶⁴⁾ has found that neutron irradiation at 790°C to 2.5×10^{22} n/cm² (E> 0.1 MeV) produced void induced swelling of approximately 1%. Such an irradiation corresponds to a dpa value of approximately 25 if a displacement energy of 36 eV is used in a Kinchin and Pease model. The swelling of 1.12% in Nb-1 Zr ion bombarded at an effective temperature of approximately 705°C to 26 dpa agrees quite well with Wiffen's values although there is a

considerable difference in the size and number density of voids in the two studies. In general, the voids were fewer, but larger in the neutron irradiated samples.

The apparent temperature threshold for void formation in the present studies is at least 560°C for pure Nb and between 525 and 560 for Nb-1 Zr. This agrees with Nb which had been neutron irradiated to approximately 3 dpa at 430°C⁽⁶²⁾ in which only loops, no voids have been found. However, Wiffen⁽⁶⁵⁾ has reported that voids do form in pure Nb at 425°C when neutron irradiated to approximately 25 dpa. It is quite possible that the difference could be due to the neutronically produced helium atoms. Even though helium is not essential for void nucleation, it may have an effect on the temperature threshold for void formation. Since future CTR materials will contain several hundred ppm He atoms, this threshold effect should be thoroughly investigated.

It is also difficult to compare the present results for V to neutron irradiated material because the lack of data. Again, Wiffen⁽⁶⁶⁾ has conducted the most complete study of voids in V and found that voids form over the range from 385°C to 600°C when irradiated to 1.4×10^{22} n/cm² (E > 0.1 MeV).^{*} He reported that no voids could be formed during 660 and 710°C irradiations. This upper temperature limit agrees quite well with our results when voids could be found after bombardment at an effective temperature of 592°C but not after 656°C. It is interesting to note that the thin ribbon precipitates noted after the effective bombarding temperature of 526°C (Figure J.6) was also observed by Wiffen⁽⁶⁶⁾ in

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^{*} This exposure corresponds to approximately 10 dpa.

vanadium which contained approximately 1000 ppm of interstitial impurities. The V used for the present study contained approximately 1100 ppm of interstitial impurities. The similarity in microstructures is especially striking considering that the neutron irradiation took several months while the identical structure was produced by ion bombardment in only a few minutes.

The morphology of voids in Nb after the 1000°C bombardment was expected to be somewhat the same as that reported for bcc Mo after high temperature neutron irradiation, but the shapes turned out to be slightly different. Rau, et al⁽⁶⁷⁾ and Kulcinski, et al⁽⁶⁸⁾ found that the void morphology in neutron irradiated Mo is consistent with {110} octahedra with {100} truncations. The situation is somewhat reversed in Nb where the voids take the shape of {100} cubes with {110} truncation (see Figure J.4). It is quite possible that the rate at which defects are produced in the metals may alter the morphology, and studies of higher temperature irradiated Mo may provide more information on this point. The cubic voids in V are consistent with observations by Elen⁽⁷³⁾ who found the voids to be bounded by {100} planes.

The alignment of the voids in Nb is similar to ion bombardment results on Ni⁽⁴⁶⁾, Mo^(36,70) and neutron irradiation results on Mo, Nb and Ta.⁽⁶⁴⁾ The fact that the alignment occurs in both types of studies which have widely differing defect production rates establishes that such structures do not solely result from ion bombardment. Also the lack of inert gas atoms in the ion bombardment experiments rules out inert gas nucleation phenomena. It has been suggested⁽⁷¹⁾ that the alignment of voids is due to the elastic anisotropy of the materials. The anisotropy ratio, $2C_{44}/(C_{11} - C_{12})$, is

2.52, 0.78 and 0.51 for Ni, Mo and Nb, respectively.⁽⁷²⁾ (A value of 1 is completely isotropic.) The exact mechanism by which such anisotropy might cause the voids to form the aligned structures is not known at this time.

Conclusions

The general conclusions of this study are:

- Microstructures and temperature thresholds for voids formed by a few minutes of 7.5 MeV Ta ion and 5 MeV Ni ion bombardment are similar to those produced by few months of neutron irradiation.
- Significant swelling and swelling gradients will exist in Nb, Nb-1 Zr, Mo, TZM, or V structural components exposed to only a few weeks of anticipated CTR neutron damaged conditions.
- Inert gas atoms such as helium are not required to nucleate voids in Nb, Nb-1 Zr, Mo, TZM or V.

FIGURE J.1 Displaced Atom Density for Ta and Ni Ions in Mo Metal

Mo

Nb

900°C

1000°C

FIGURE J.2 Voids in Mo and Nb After 5 MeV Ni Bombardment at 900°C and 1000°C

DAMAGE ~ 5 dpa

FIGURE J.3 Effect of Bombardment Temperature on the Production of Voids in TZM

FIGURE J.4 Void Induced Swelling in Refractory Metals After Bombardment to Approximately 5 dpa

FIGURE J.5 Cubical Voids in Niobium After 1000°C Ion Bombardment to 5 dpa

FIGURE J.6 Alignment of Voids in Niobium After 800°C Bombardment to Approximately 5 dpa

700°C

800°C

FIGURE J.7 Voids in Vanadium After 7.5 MeV Ta Ion Bombardment

700°C

800°C

900°C

FIGURE J.8 Effect of Irradiation Temperature on the Formation of Voids in Nb-1 Zr.
Damage Level Approximately 30 dpa

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