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TRANSMUTATION OF MASSIVE LOADINGS OF ^{137}Cs IN THE BLANKET OF A CONTROLLED THERMONUCLEAR REACTOR

B. F. Gore†

B. R. Leonard, Jr.

Battelle - Pacific Northwest Laboratory
Richland, Washington 99352

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DISCLAIMER

ABSTRACT

Calculations have been performed which indicate the possibility of reducing below ten years the effective half-life for transmutation of massive loadings of ^{137}Cs placed in a CTR blanket. The blanket studied is a modification of the cylindrical "standard blanket" calculational benchmark, having 1 cm of Nb wall material followed by a 20 cm target zone, 70 cm of moderator and a 6 cm absorber of Li. The "standard blanket" source yielding a wall loading of 10 MW/m^2 of 14 MeV neutrons is assumed. Neutron production by (n,2n) reactions in a Be moderator is used to increase the thermal flux. For an 80% target zone loading of ^{137}Cs, a transmutation rate of 290 kg per year per meter of CTR length is obtained, with a half-life (including radioactive decay) of 9.9 years. At this loading, the transmutation rate in roughly one percent of the length of a CTR blanket would balance the production rate in a fission reactor of the same power.

INTRODUCTION

Calculations have been performed to study the feasibility of transmuting fission products by neutron-induced reactions in the blanket region of a controlled thermonuclear fusion reactor (CTR). More specifically, attention was focused upon the possibility of significantly shortening the 30 year half-life of ^{137}Cs by this process. Because ^{137}Cs is copiously produced in both plutonium and uranium fissions, accelerating its decay could help to ease the radioactive waste problems attendant upon nuclear power generation. Further, because it has the lowest thermal neutron capture cross section of the moderately long-lived fission products, ^{137}Cs represents a "worst case" for this technique.

Because of the very low neutron capture cross section of ^{137}Cs, a high thermal neutron flux is required to transmute it at a rate equal to its natural rate of radioactive decay. Specifically, an integrated $1/v$[1] flux of 7×10^{15} n-cm^{-2} - sec^{-1} is required. Initial calculations were performed using source and blanket configurations related to the "standard blanket" calculational benchmark defined at the ORNL Fusion Technology Conference.[2] These calculations showed that flux traps could be designed to produce $1/v$ fluxes exceeding 10^{16} n-cm^{-2} - sec^{-1}. Consequently, interesting transmutation rates could be expected for infinitely dilute loadings of ^{137}Cs.

In order for transmutation to be useful for the reduction of fission product inventories, it must be possible to transmute significant quantities of target material at interesting rates. Therefore, calculations were performed to study the transmutation rate for macroscopic loadings of ^{137}Cs in a model CTR blanket. The results of these calculations indicate that a blanket region can be designed in which ^{137}Cs is transmuted more

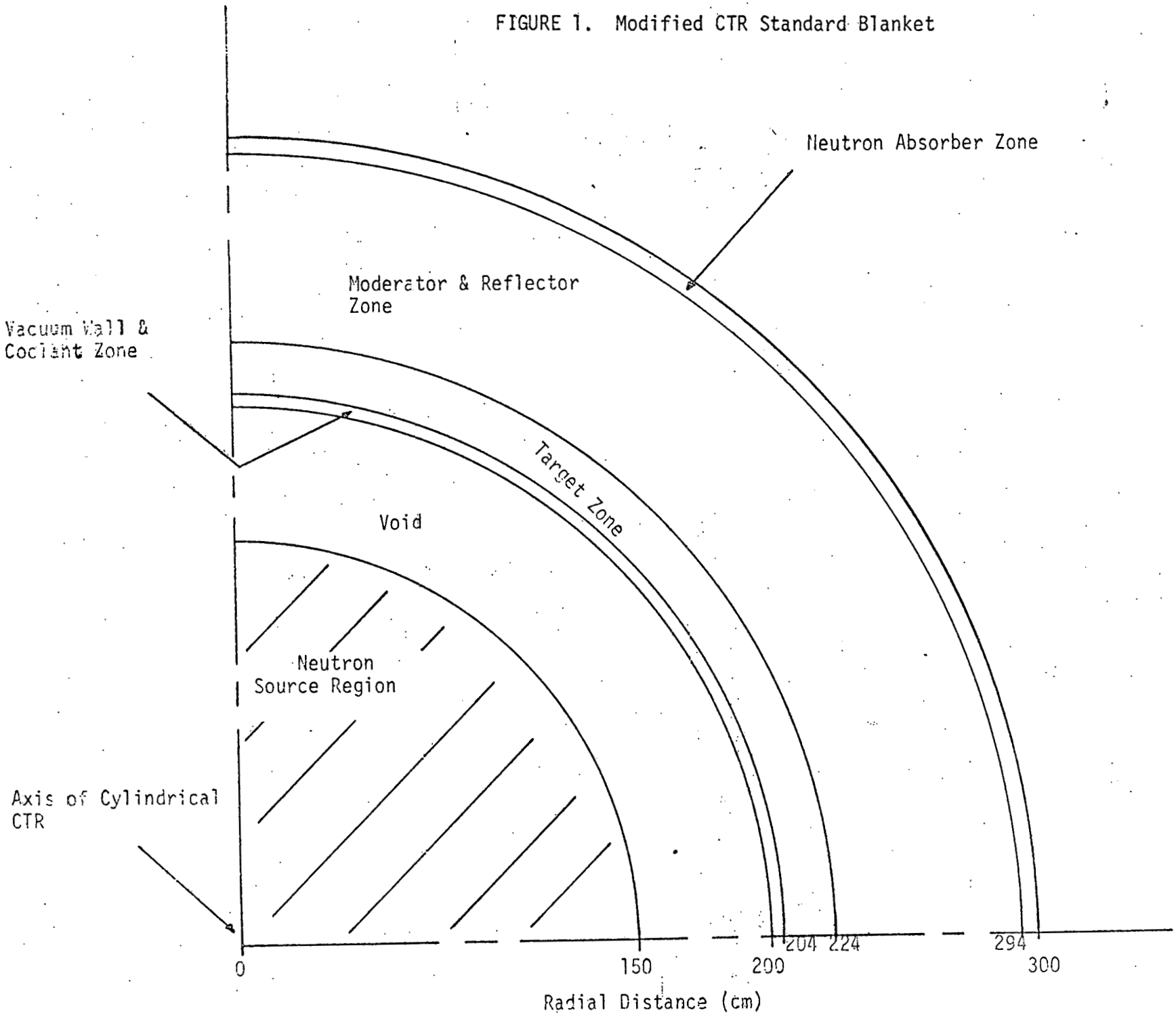
than twice as fast as it decays naturally, even at loadings exceeding 50% by volume. At such loadings the transmutation rate in roughly 1% of the blanket region of a CTR would balance the production rate in a fission reactor producing the same power.

CALCULATIONS AND RESULTS

The CTR "standard blanket"[2] is a cylindrical annulus (perhaps a section through a torus), with inner and outer radii of 200 and 300 cm respectively. Its functions are to absorb the kinetic energy of the 14 MeV fusion neutrons, breed tritium, and shield the outer (presumably superconducting) magnets from neutrons. The blanket zones are cylindrical annuli, starting with an inner Nb vacuum wall 0.5 cm thick. This is followed by a 3 cm layer of Li "mix" (94% Li, 6% Nb) which represents a coolant zone. A 0.5 cm Nb wall separates this from a second zone of Li mix which is 60 cm thick. This is followed by a graphite reflector 30 cm thick and a final zone of Li mix 6 cm thick. The specified DT fusion neutron source of the "standard blanket" is a 14 MeV neutron current at the vacuum wall of 4.43×10^{14} n-cm^{-2}-sec^{-1} corresponding to a neutron energy transport of 10 MW-m^{-2}. All calculations reported herein were normalized to this wall loading of fusion neutrons.

The concept explored in these calculations is to replace the central layer of Li mix with pure moderator as shown in Figure 1, to produce a high thermal flux. Because neutronics calculations of the standard blanket have indicated a tritium breeding rate in excess of 1.25,[3] adequate tritium supplies for the CTR are assumed available. Tritium production is ignored here, except as a by-product of the outer neutron-absorbing layer of Li mix which protects the magnets from neutrons leaking through the

FIGURE 1. Modified CTR Standard Blanket



moderator. (Due to more effective moderation of fast neutrons this blanket reduced neutron leakage to the magnets by at least a factor of four below levels calculated for the standard blanket.)

Using the ANISN code[4] the initial calculations explored ways to produce a high 1/v flux in the moderator region. Reactor density graphite was tried first; then beryllium was tried for its better moderating properties and its low threshold energy for neutron production through (n,2n) reactions. In the calculation for Be the reflector zone was also filled with Be. Both moderator and reflector zones were loaded to an atom density of 65% that of metallic Be. These calculations indicated considerable neutron losses to the inner coolant layer of Li mix. Consequently, it was decided to postulate gas cooling and remove the mix from that zone. The results of both sets of calculations are listed in Table I, and indicate the possibility of achieving high average fluxes over an extensive region of moderator.[5]

In addition to neutron capture, ^{137}Cs will undergo transmutation to the stable isotope ^{136}Ba in the high-energy neutron flux via the (n,2n) reaction to ^{136}Cs followed by rapid (13 d) beta decay. The calculated fast neutron flux near the inner edge of the blanket plus an assumed (n,2n) cross section of 1.5 b would give an additional infinitely dilute specific activity of 0.021 per atom per year. Even though the high energy neutron flux decreases rapidly with penetration into the moderator, this process should enhance transmutation rates significantly if fission product loadings are confined to the inner portion of the blanket (which is also the high thermal flux region). Consequently, further calculations using model ^{137}Cs cross sections were undertaken.

TABLE I. Integrated 1/v flux calculated for modifications of the CTR "standard blanket" discussed in the text. Also shown are specific activities calculated using the ^{137}Cs thermal neutron capture cross section. Averages are over the blanket region between radii 204 and 264 cm. The ^{137}Cs specific activity due to radioactive decay is 0.023 per year.

	$\phi_{1/v}$ (cm^{-2}-sec^{-1})		$<\sigma\phi>$ (per year)	
	Max.	Ave.	Max.	Ave.
Graphite				
w/Li Shell	2.8×10^{15}	1.7×10^{15}	0.010	0.006
wo/Li Shell	7.6×10^{15}	4.7×10^{15}	0.026	0.016
Beryllium				
w/Li Shell	7.4×10^{15}	4.3×10^{15}	0.026	0.015
wo/Li Shell	2.2×10^{16}	1.1×10^{16}	0.076	0.038

The ENDF/B-III description of ^{137}Cs (MAT 1230) is not adequate to perform the neutronics calculations. This description not only does not contain an (n,2n) cross section, but all of the calculated non-elastic cross section other than capture is attributed to inelastic scattering. Therefore, in order to obtain a reasonably valid neutron transport calculation the microscopic cross sections of the nearby nucleus ^{133}Cs, as given on ENDF/B-III (MAT 1141)[6] were used to build a model for ^{137}Cs cross sections. The ^{133}Cs description contains (n,p), (n,α) and (n,2n) reactions in addition to capture, and elastic and inelastic scattering. In order to build a ^{137}Cs cross section file only the ^{137}Cs capture cross section of MAT-1230 was substituted for the ^{133}Cs capture file.[7]

The calculated specific activities for neutron capture on ^{137}Cs are due almost entirely to the thermal group. There has apparently been only one determination of the ^{137}Cs thermal capture cross section. This measurement was the cross section to produce the 32 min ^{138g}Cs activity. The value obtained was 0.110 ± 0.050 b.* Thus the calculated neutron capture rates are uncertain by some fifty percent due to the uncertainty in the cross section.

Calculations were performed for various loadings of the model ^{137}Cs in the blanket for the most favorable case of the initial study - the Be moderated case with a Be reflector, with the inner Li layer replaced by

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* The error of ± 0.033 b quoted in BNL 325, 2nd Ed., Suppl 2, Neutron Cross Sections Vol II B was only the random error assigned by Stupegia.[8] Stupegia estimated the total error to be the value given in the text.

void. For these calculations the atom density of Be was taken to be 80% of metallic, with void space assumed for gas cooling. Figure 2 presents the results of two studies in which the Cs was loaded into the inner 20 cm of blanket (between radii of 204 and 224 cm). In one study, the region was filled to 80% by volume, with Cs and Be, in varying proportions. In the other study, only Cs in three different volume fractions was placed in this region.

In order to understand the calculated results shown on Figure 2, it should be recognized that while the mass densities of elemental Be and Cs differ by less than 2%, the atom density of Cs is less than 7% that of Be. One result of this is that, on a volume percent basis, Be is a much better moderator. Another result is that even a full volume loading of pure Cs is so dilute that there is little attenuation of the high energy neutron flux. Thus, when only Cs is present in the target zone the (n,2n) reaction probability is weakly dependent upon Cs concentration. When Be is loaded with the Cs, however, the decrease in Cs (n,2n) reaction probability with increasing Be loadings is rapid.

In the cases where the target zone was filled to 80% by volume with varying Cs/Be fractions, the systematic decrease in the Cs (n,γ) absorption probability with increasing Cs fraction follows from the decreasing ability of the mixture to attenuate the high energy neutron flux. The radius at which the thermal flux reaches a maximum increases with increasing Cs fraction, decreasing the probability of (n,γ) absorption per Cs atom.

Table 2 lists both average and maximum reaction rates in the target zone and the corresponding effective half lives (including radioactive

FIGURE 2. ^{137}Cs TRANSMUTATION PROBABILITY PER YEAR

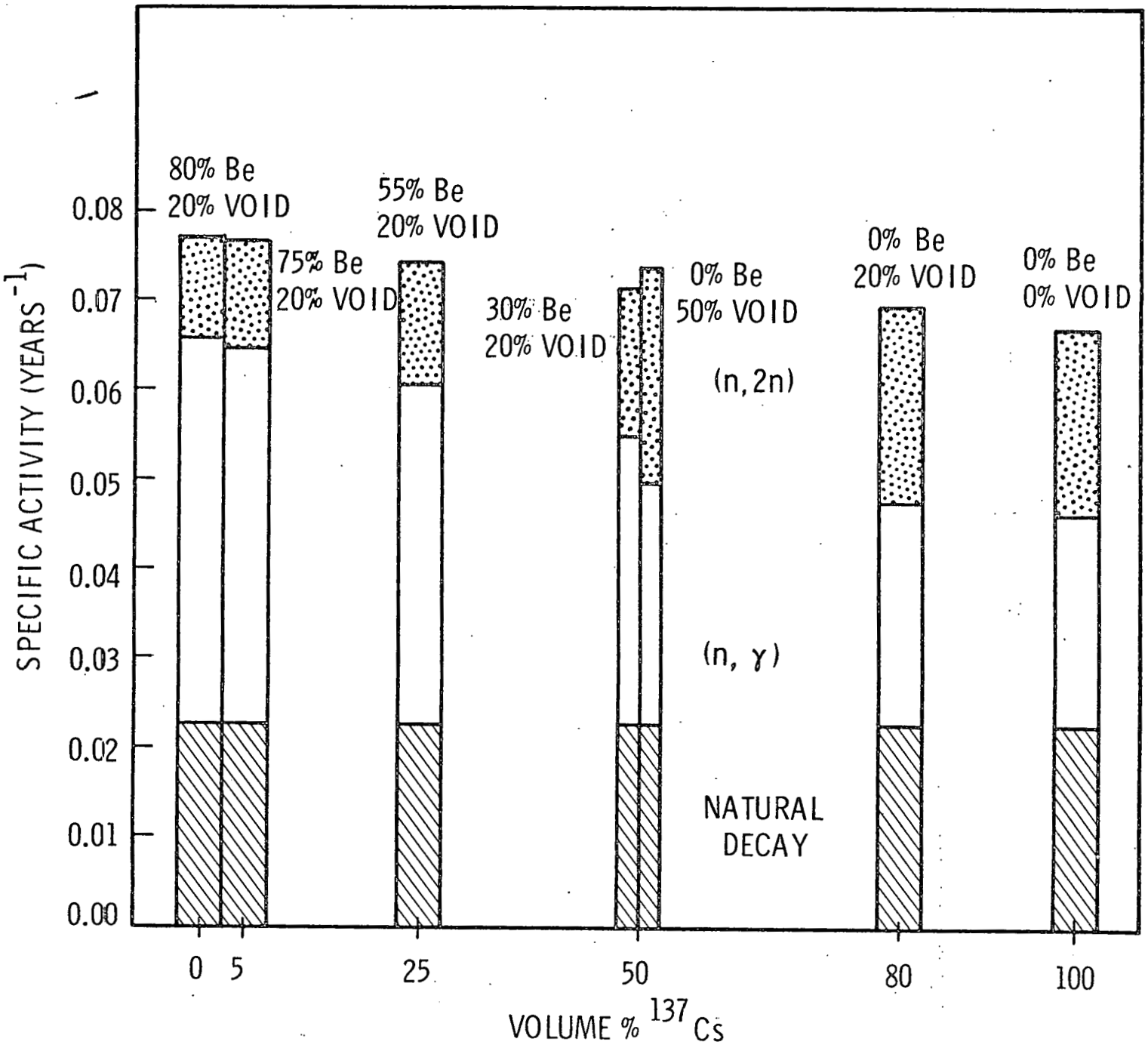

TABLE 2. Calculated Transmutation Rates of Cs-137 in the Blanket of a Tokomak Fusion Reactor†

Volume Fraction %Cs/%Be	Average $\phi(1/v)\times10^{-16}$ (Neut/cm^2-sec)	Average Reaction Rate $\langle\sigma\phi\rangle_{n,\gamma}$ fraction (year)	Average Reaction Rate $\langle\sigma o\rangle_{n,2n}$ fraction (year)	Average Total Effective T-1/2* (years)	Average Total Transmutation Rate* (kg-m^{-1}-yr^{-1})	Maximum $\phi(1/v)\times10^{-16}$ (Neut/cm^2-sec)	Maximum Reaction Rate $\langle\sigma\phi\rangle_{n,\gamma}$ fraction (year)	Maximum Reaction Rate $\langle\sigma\phi\rangle_{n,2n}$ fraction (year)	Maximum Total Effective T-1/2* (years)
C/80	1.19	0.0428	0.0115	9.0	0	1.26	0.0450	0.0266	7.3
5/75	1.16	0.0419	0.0119	9.0	20	1.23	0.0440	0.0267	7.4
25/25	1.05	0.0379	0.0137	9.3	95	1.13	0.0402	0.0273	7.6
50/30	0.874	0.0321	0.0168	9.7	185	0.958	0.0349	0.0281	8.1
50/0	0.743	0.0275	0.0241	9.3	190	0.763	0.0282	0.0292	8.6
80/0	0.671	0.0250	0.0221	9.9	290	0.702	0.0262	0.0290	8.9
100/0	0.633	0.0238	0.0209	10.3	350	0.668	0.0252	0.0289	9.0
100/0††	1.29	0.0470	0.0268	7.2	500	1.31	0.0478	0.0387	7.0

*Includes radioactive decay activity of 0.0229 per atom per year.

†This device produces 200 MW(t) per meter of CTR device length.

††Nb wall material (1 cm thick) removed.

decay). Also shown are the integrated 1/v fluxes and the total transmutation rates per meter of CTR device length. The bottom row of this table lists the results of calculations for a case in which the Nb wall material is removed. It is included to indicate the upper limits of improvement obtainable by redesign of the inner wall region.

Since the inclusion of void space for coolant will doubtless be necessary in the target zone, 80% volume fraction loading of pure Cs in the target zone with the Nb walls in place may be the most realistic case. The calculated transmutation rate of 290 kg per year per meter of device length for this case is such that transmutation in roughly 1% of the length of a CTR blanket would balance the production rate in a fission reactor of the same thermal power. This transmutation rate, with an effective half-life of 9.9 years, is deemed interesting.

Beryllium has not been widely used as a moderating material in nuclear fission reactors. This is apparently due to the manufacturing expense related to Be toxicity, and due to the poor structural stability of Be under neutron irradiation. Consequently, calculations have been made to determine the extent to which the Be content of the blanket can be replaced by graphite without losing the flux enhancement obtained through Be(n,2n) reactions. These calculations were carried out for the case with a 25% Cs, 55% Be loading in the target zone. No change in the Cs transmutation rate was found when the outer 50 cm of Be was replaced by graphite; replacement of the entire 70 cm Be zone external to the target zone decreased the $Cs(n,\gamma)$ transmutation rate by less than 10%. Thus large quantities of Be in the blanket may not be necessary to achieve the desired neutron multiplication.

A second result of lesser importance was also obtained in studying the combination Be-graphite moderator. The tritium production per source neutron due to $^{6}Li(n,\gamma)$ capture reactions in the outer absorber layer increased from 0.18 for the pure Be moderator case to 0.42 when all Be external to the target zone was replaced by graphite. This indication that tritium production may be achieved simultaneously with transmutation led to a calculation for which the target zone was followed by 20 cm of Be, and the remaining 56 cm of the blanket was filled with the Li-Nb absorber mix. Tritium production increased to 0.94 per source neutron with a decrease of only 20% in the $Cs(n,\gamma)$ transmutation rate below the rate for the full Be blanket case. Thus, optimization of both tritium production and transmutation rates seems possible.

Other aspects of the relative merits of beryllium and carbon as moderators in CTR blankets include the gas production rates, which strongly determine structural stability, and the induced radioactivity. These effects might be expected to differ in the 14 MeV neutron source flux of the CTR from the same effects in a fission reactor neutron flux. Consequently, these relative effects have been estimated for the DT fusion reactor.

Gas Production

The neutron multiplication of the beryllium blanket through the (n,2n) reaction is achieved at the expense of helium gas production via the reaction:

$$^{9}Be(n,2n)\ ^{8}Be \xrightarrow{10^{-16}s} 2\ ^{4}He.$$

With the neutron spectra produced by the 14 MeV DT source, additional helium and tritium gas are also produced by means of the reactions

$$^{9}Be(n,^{4}He)\ ^{6}Li$$

$$^{9}Be(n,^{3}H)\ ^{7}Li$$

and the secondary reaction

$$^{6}Li(n,^{3}H)\ ^{4}He.$$

Helium is produced in the graphite blanket by the reactions:

$$^{12}C(n,\ ^{4}He)\ ^{9}Be$$

$$^{12}C(n,n')\ 3\ ^{4}He.$$

Calculations, which are reported elsewhere,[9] have been performed for 100 cm blankets of pure Be and pure graphite at 80% theoretical density. The production rates of helium and tritium in the first cm of a Be blanket, given in terms of percent of initial blanket atoms per year, are 4.5% yr^{-1} and 0.2% yr^{-1}, respectively. In a graphite blanket the corresponding helium production rate is 2.2% yr^{-1}.

The calculated helium production rates in graphite are about half of the beryllium values. Nevertheless, the gas production in the beryllium blanket is actually less than in the graphite per thermal neutron available for transmutation. In any event, the gas production rates in both materials are so high that neither material would have any structural stability. To be used in a 14 MeV DT neutron flux of this magnitude either material would have to be encased in a structural material with a greater resistance to radiation damage.

Induced Radioactivities

The activities of the radioactive daughters produced in the same blanket have also been estimated. In beryllium the only long-term activity is the 1.6×10^6 yr beta emitter ^{10}Be produced by neutron capture. Calculations indicate a buildup per day of 5.4 curies per meter of length for the full blanket thickness. In graphite, ^{10}Be activity is also produced by the (n,α) reaction on the 1.1% ^{13}C isotope. Since there are no experimental determinations of this cross section, a value was obtained for this activity by assuming that the $^{13}C(n,\alpha)$ cross section was equal to the sum of the $^{12}C(n,\alpha)$ and $^{12}C(n,3\alpha)$ cross sections. Thus the value, 0.015 curies per day per meter, may be erroneous by a factor of several. This is not of particular importance since the dominant activity in graphite is that of the 5730 yr beta emitter ^{14}C produced by neutron capture by ^{13}C. This ^{14}C production rate is initially 0.83 curies per day per meter. It is actually time dependent since there will be appreciable buildup of ^{13}C during irradiation by neutron capture in ^{12}C. However, this will be compensated in part by the (n,2n) reaction on ^{13}C which has a threshold energy of 5.3 MeV. The analysis of the effects of these secondary reactions has not been carried out. These calculated values of activity do not indicate any particular problems.

Thus, interesting transmutation rates may be obtained for massive loadings of ^{137}Cs in a CTR blanket. Transmutation of significant quantities of Cs appears feasible at these interesting transmutation rates. Central to this achievement is the inclusion of Be in the inner blanket regions, although large amounts may not be necessary. Problems associated with the use of Be as a moderator appear not to be much worse than similar ones associated with the use of a graphite moderator.

REFERENCES AND FOOTNOTES

1. $$\phi_{1/v} = E_{TH}^{1/2} \int_{E_{TH}}^{E_{MAX}} E^{-1/2}\, \phi(E)\, dE$$

2. International Working Sessions on Fusion Reactor Technology, ORNL June 28-July 3, 1971, CONF-710624.

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4. W. W. Engle, Jr., A User's Manual for ANISN, A One Dimensional Discrete Ordinates Transport Code with Anisotropic Scattering, K-1693, March 30, 1967.

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   Calculations were performed using the ANISN code in the P3-S8 Approximation. Cross sections, originally processed to one hundred energy group format by the ETOG code, were "collapsed" to twenty-seven groups (using ANISN) by flux-weighted averaging over an infinite carbon spectrum to reduce computation time.

5. An input error, discovered after calculations were complete, caused the Nb atom density in the two Nb wall regions to be only 6% of that of Nb metal. Consequent underprediction of neutron captures in Nb causes the fluxes listed in Table I to be somewhat optimistic. Recalculation of the most favorable case (Be moderated, no inner Li shell) showed reductions of almost 50% in average and maximum flux values. The calculations presented in the remainder of this paper were done with the Nb shells at metallic density.

   This unpremeditated parametric study indicates that improved transmutation rates may be obtainable by redesigning this region paying careful attention to the neutronic properties of the structural materials.

6. ENDF/B-DLC-2 B Cross Section Data, Radiation Shielding Center, Oak Ridge National Laboratory.

7. 100-Group Neutron Cross-Section Data Based on ENDF/B, Radiation Shielding Center, Oak Ridge National Laboratory

The ^{133}Cs one hundred energy group cross sections and transfer matrix were obtained from the RSIC tape library and "collapsed" by the ANISN code into 27 group structure as discussed in footnote 5. The capture cross sections of ^{137}Cs and ^{133}Cs were plotted from ENDF/B listings and 100 group cross sections obtained from the graphs. For each of the 100 energy groups the ^{133}Cs capture cross section was subtracted from σ_{total} and $\sigma_{absorption}$ and the ^{137}Cs capture cross section added. The resulting 100 group cross sections, and the ^{137}Cs capture cross sections were "collapsed" to 27 group format by hand over the same flux spectrum used in the ANISN collapse. σ_{total}, $\sigma_{absorption}$ and $\sigma_{capture}$ in the collapsed ^{133}Cs cross section set were replaced by the resulting values, yielding a model cross section set for ^{137}Cs. The other cross sections used in these calculations were obtained from ENDF/B-III.

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9. W. C. Wolkenhauer, B. R. Leonard, Jr., and B. F. Gore, Transmutation of High Level Radioactive Waste with a Controlled Thermonuclear Reactor, BNWL-1772, July 1973.