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## Fission Multipliers for D-D/D-T Neutron Generators

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**Abstract**— A compact D-D/D-T fusion based neutron generator is being designed at the Lawrence Berkeley National Laboratory to have a potential yield of  $10^{12}$  D-D n/s and  $10^{14}$  D-T n/s. Because of its high neutron yield and compact size (~20 cm in diameter by 4 cm long), this neutron generator design will be suitable for many applications. However, some applications required higher flux available from nuclear reactors and spallation neutron sources operated with GeV proton beams. In this study, a subcritical fission multiplier with  $k_{\text{eff}}$  of 0.98 is coupled with the compact neutron generators in order to increase the neutron flux output. We have chosen two applications to show the gain in flux due to the use of fission multipliers – in-core irradiation and out-of-core irradiation. For the in-core irradiation, we have shown that a gain of ~25 can be achieved in a positron production system using D-T generator. For the out-of-core irradiation, a gain of ~17 times is obtained in Boron Neutron Capture Therapy (BNCT) using a D-D neutron generator. The total number of fission neutrons generated by a source neutron in a fission multiplier with  $k_{\text{eff}}$  is ~50. For the out-of-core irradiation, the theoretical maximum net multiplication is ~30 due to the absorption of neutrons in the fuel. A discussion of the achievable multiplication and the theoretical multiplication will be presented in this paper.

### I. INTRODUCTION

The compact D-D/D-T fusion neutron generator currently under development by the Ion Beam and Technology (IBT) Program at Lawrence Berkeley National Laboratory has an expected yield of  $10^{12}$  and  $10^{14}$  n/s respectively. The high neutron yield is envisioned because of its innovative cylindrical shaped RF driven ion source. The ions are extracted radially toward a cylindrical target around the source. This geometric arrangement allows a large target area with a small source size.

The high neutron yield makes this new generation of neutron generator suitable for a wide variety of applications for which those conventional neutron generators are unable to provide sufficient flux. As some scientists and engineers are building more powerful neutrons sources such as highly enriched uranium (HEU) fueled research reactors and spallation neutron sources for applications that require higher neutron flux, people also try the approach of increasing the neutron flux from neutron generators by a fission multiplier. The idea of using a fission multiplier is not new; its concept is similar to those accelerator driven systems (ADS) for transmutation of nuclear waste. Instead of driving the subcritical pile by a spallation neutron source to transmute hazardous nuclear waste, the D-D/D-T neutron driven fission multiplier is targeted to applications that require a neutron yield in the range of  $10^{14}$  to  $10^{16}$  n/s. The feasibility of using a D-D/D-T fusion neutron driven fission multiplier is studied for two types of applications in this paper.

One application of thermal neutrons is to induce prompt  $\gamma$ -rays by the capture process in  $^{113}\text{Cd}$  so that the  $\gamma$ -rays from the capture process can produce positron by the pair production process.<sup>1</sup> The system for positron production described in Reference 1 is consisted of a  $^{113}\text{Cd}$  cap with a dimension of 6.5 cm diameter by 6.5 cm long with platinum plates inside. On the  $^{113}\text{Cd}$  cap, thermal neutrons are captured to produce high-energy  $\gamma$ -rays. The platinum foils inside the  $^{113}\text{Cd}$  cap generate positrons by stopping some of these  $\gamma$ -rays with the pair production process. The platinum foils also act as a positron moderator because of its negative work function of -1.8 eV in polycrystalline Pt.<sup>2</sup> In this study, the  $^{113}\text{Cd}$  cap is placed very closed to a subcritical pile driven by a D-T neutron generator. The discussion presented in this paper is limited to the parameters that affect the optimization of thermal neutron absorption rate in the  $^{113}\text{Cd}$  cap.

The other application being investigated is a subcritical multiplier driven by a D-D neutron generator with a yield of  $10^{12}$  n/s for Boron Neutron Capture Therapy (BNCT). Previously, a D-T neutron source with a yield of  $10^{14}$  n/s is shown to be adequate to give a treatment time of ~45 minutes.<sup>3</sup> However, there is an interest to use a subcritical pile to boost the neutron yield from a D-D neutron generator due to the public awareness and concerns of using tritium in a hospital environment. Unlike fission converter plates that convert the thermal neutrons in a research reactor to fast neutrons, a subcritical fission multiplier is designed to increase the flux of a neutron source. Because most neutrons produced in a reactor are thermalized when they get out of the reactor core, it is necessary to convert these thermal neutrons into fast neutrons



and re-moderate them to epithermal.<sup>4</sup> A fission converter assembly with a  $k_{eff}$  of 0.26 was shown to be capable of producing  $9 \cdot 10^9$  n/cm<sup>2</sup>s in a 10 MW research reactor. A  $k_{eff}$  of 0.26 would be sufficient for the fission converter plates because they are irradiated with a high thermal neutron flux. A fission multiplier, on the other hand, usually requires a  $k_{eff}$  close to 1 because its goal is to produce more neutrons. A  $k_{eff}$  of 0.98 was chosen in this study.

These two chosen applications can be viewed as two different categories: (a) in-core irradiation and (b) out-of-core irradiation. For in-core irradiation, the sample being irradiated is placed inside or close to the vicinity of the multiplier assembly. For example, the <sup>113</sup>Cd cap of a positron production system is located very close to the fuel. Other examples of in-core irradiation are radioisotope production and Instrumental Neutron Activation Analysis (INAA). For out-of-core irradiation, the neutron beam is extracted out of the multiplier assembly. Examples of this category include neutron radiography, Prompt Gamma Neutron Activation Analysis (PGAA) and etc. BNCT is chosen for this category in this study because it requires an epithermal neutron beam that is not readily available without contaminations from thermal and/or fast neutrons in most reactors.

## II. SOURCE BRIGHTNESS AND NET MULTIPLICATION

The major factors that forbid a fission multiplier from improving the neutron flux are source brightness and the fuel material.

Any neutron source can be designed to produce a lot of neutrons. However, a high neutron yield alone does not represent how useful these neutrons are. Flux is defined as the number of neutrons crossing a unit area per unit time. It can also be viewed in the following way:

$$\Phi = nv \quad (1)$$

$n$  is the neutron density,  
 $v$  is the velocity of neutron.

A neutron source with a high yield must also have a high neutron density in order to provide a high neutron flux. When the neutron generator is coupled with a neutron generator, the volume of the source increases. As a result, the neutron density increases slower than the yield and the neutron flux will not be improved linearly with the true system multiplication factor. The definition of "true system multiplication" taken from the user manual of MCNP is:<sup>5</sup>

$$1 + k_{eff} + k_{eff}^2 + \dots + k_{eff}^n + \dots = \frac{1}{1 - k_{eff}} \quad (2)$$

The true system multiplication includes the neutrons that undergo the fission process. Due to its naming convention, people may misinterpret that an optimized fission multiplier has a net gain in neutron flux equal to the true system multiplication if the source brightness is not reduced in consequence of introducing a subcritical pile. The net multiplication,  $M$ , for a fixed source problem in MCNP is defined as:

$$M = 1 + G_f + G_x = W_e + W_c \quad (3)$$

$G_f$  is the weight of gain from fission,  
 $G_x$  is the weight of gain from n,xn reaction,  
 $W_e$  is the weight of escape from the system,  
 $W_c$  is the weight of capture in the system.\*

$$G_f = (\nu - 1)W_f \quad (4)$$

$W_f$  is the weight of fission.

Equations 3 and 4 explicitly show that the net multiplication takes account of the neutrons undergo fission and capture in the system. If  $M$  is fixed,  $W_e$  increases when  $W_c$  decreases. For a system with a  $k_{eff}$  of 0.98,  $M$  is approximately 30 and 33 for a core loaded with 100% <sup>235</sup>U and a core loaded with reactor-grade Pu only. There is a minimum  $W_c$  even in a perfect system because neutrons are absorbed in the fuel by other process. If the loss due to source brightness and the absorption in moderator and other structure materials is negligible, the maximum  $W_e$  strongly depends on  $M$  and  $W_c$  of the fuel.

## III. DESIGNS OF FISSION MULTIPLIERS

From the discussion in Section II, several key issues in optimizing a subcritical fission multiplier are identified:

1. The fuel should have a high content of fissile material in order to reduce the absorption in the fuel. (i.e. no <sup>238</sup>U)

\*  $W_c$  includes all absorption process in a fixed source problem except the fission process.



2.  $\bar{\nu}$ , the number of fission neutrons per fission, for the fissile material should be large in order to minimize the number of neutrons that undergo fission.
3. The size of the fission multiplier should be compact with high leakage toward the sample being irradiated. In other words, the sample should see a fuel assembly that is optically thin.
4. The moderator and other structure materials should not have large absorption cross-sections so that the neutrons born in the fuel can escape the core.

### III.A. In-Core Irradiation – Positron Production System

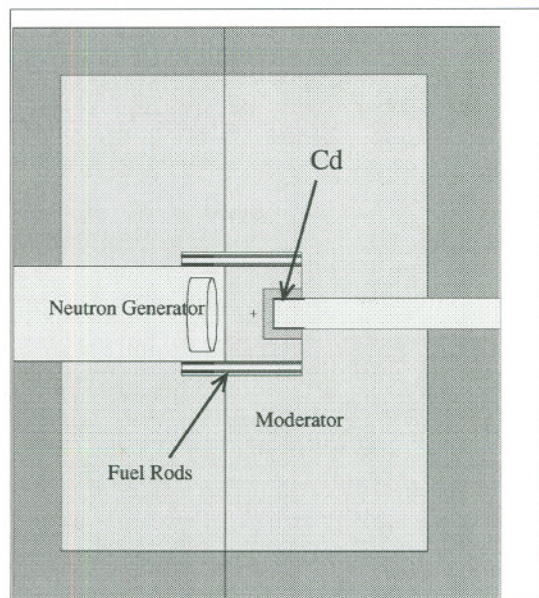
Because plutonium has a larger  $\bar{\nu}$  than uranium, the fuel chosen in this study is Pu-10Zr. Reactor-grade Pu with an isotope composition of 2%  $^{238}\text{Pu}$ , 56%  $^{239}\text{Pu}$ , 26%  $^{240}\text{Pu}$ , 11%  $^{241}\text{Pu}$  and 5%  $^{242}\text{Pu}$  by weight is used. Metallic fuel is chosen for its high Pu density. Table 1 shows other specification for the fuel rod. The total Pu loading of the subcritical assembly is 18.7 Kg.

**Table 1. Specification for the fuel and clad used in D-T driven fission multiplier**

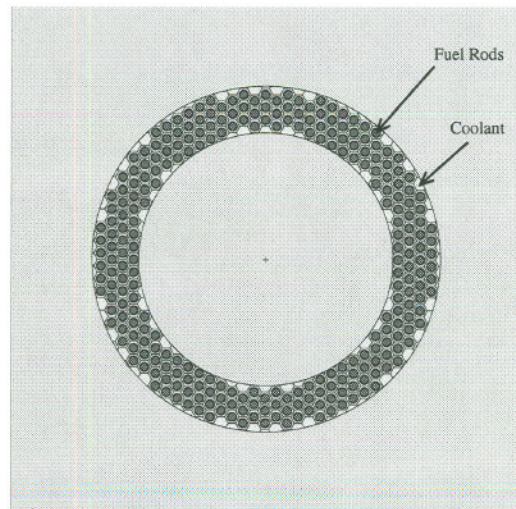
Fuel	Metallic Pu-10Zr
Density	14.36 g/cm <sup>3</sup>
Pu density	12.9 g/cm <sup>3</sup>
Fuel temperature	755 °K
Pu composition	2% $^{238}\text{Pu}$ , 56% $^{239}\text{Pu}$ , 26% $^{240}\text{Pu}$ , 11% $^{241}\text{Pu}$ , 5% $^{242}\text{Pu}$
Diameter	0.63 cm
Length	17.25 cm
Plenum	5.75 cm
Pin arrangement	Hexagonal pitch
Pitch	1 cm
Number of pins	270
Gap	Na filled
Thickness	0.2 mm
Clad	HT-9
Density	7.7 g/cm <sup>3</sup>
Thickness	0.4 mm
Composition	84.49% Fe, 11.8% Cr, 0.51% Ni, 1.03% Mo, 0.5% Mn, 0.33% V,

0.52% W, 0.21% Si, 0.21% C
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The cladding material is chosen to be martensitic stainless steel, HT-9, for its excellent strength and swelling properties under fast neutron irradiation.<sup>6</sup> Helium at a pressure of 2.3MPa is assumed to be the coolant for the subcritical assembly due to its low absorption cross-section for neutron. Beryllium is used as moderator and reflector. A beryllium moderator is used rather than a light water moderator because the light water tends to over-thermalize the neutrons before they reach the Cd cap. A secondary light water moderator surrounding the Cd cap is used to enhance the thermal neutron flux in the Cd cap. (Figure 1)



**Figure 1. A D-T driven fission multiplier showing a Cd cap surrounded by a small light water moderator**





**Figure 2. A cylindrical fuel assembly for the D-T fission multiplier**

The coaxial neutron generator has a diameter of 20 cm and a length of 4 cm. A cylindrical fuel assembly surrounds the neutron generator and the Cd cap. Its inner diameter and outer diameter are 20 cm and 27.4 cm, respectively. As shown in Figure 2, the fuel assembly is consisted of 270 fuel pins. Each of these fuel pins has a plenum filled with sodium to allow thermal expansion of the fuel pellets.

The calculation is performed with MCNP. There are two modes of neutronics calculation in MCNP: (a) fixed source and (b) criticality. When the  $k_{eff}$  of a system is close to 1, a criticality problem has almost the same result as a fixed source problem in most cases. One history of a fixed source problem with  $k_{eff}$  close to 1 can sometimes take a long time to finish. Therefore, the calculation is performed with criticality calculation first. Then, it is verified with the fixed source calculation to account for the source neutron spectrum.

### III.B. Out-of-Core Irradiation – BNCT

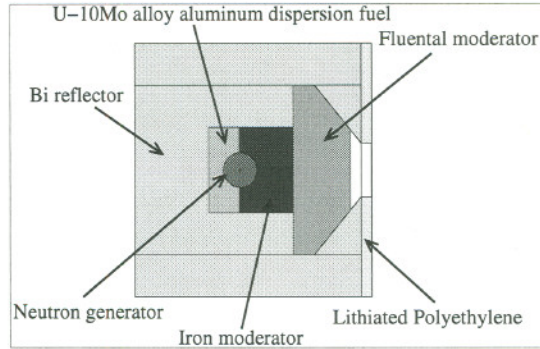
In designing a beam shaping assembly for BNCT, aluminum, iron and aluminum fluoride are some of the best moderator materials.<sup>3,7</sup> The U-Mo alloys have a very high fuel density.<sup>8</sup> The U-10Mo alloy fuel has a density of 16.8 g/cm<sup>3</sup>. The alloy fuel particles, when dispersed in an aluminum matrix (i.e. the moderator), can act as a high brightness fission neutron source. Table 2 shows some physical properties of U-10Mo alloy dispersed in an aluminum matrix. The fuel is assumed to be 100% enriched <sup>235</sup>U. Pu is not chosen here because U-10Mo shows stable behavior with respect to fuel-matrix interaction in the form of inter-diffusion.<sup>9</sup>

**Table 2. Physical properties of U-10Mo alloy aluminum matrix dispersion fuel**

Fuel composition	90% U, 10% Mo
Aluminum matrix volume (%)	71
Fuel volume (%)	29
Fuel density	4.4 g-U/cm <sup>3</sup>

Because the fuel is dispersed in the moderator, the change in moderator is minimum. The beam shaping assembly for D-D neutron source is taken from Reference 7. The back of its Fe moderator is replaced with the dispersion fuel as shown in Figure 3. A power of ~630 W is calculated for this multiplier from the neutron

yield of the D-D neutron generator and the  $k_{eff}$  of the fission multiplier. Therefore, for simplicity reason and reduction of computer time, there is no coolant channel in this computational model.



**Figure 3. A D-D driven fission multiplier with metal alloy with dispersed in its moderator**

A conical D-D neutron generator is used for the BNCT system. The conical neutron generator is another designs under development at LBNL. It has a diameter of 16 cm and a length of 32 cm. The iron moderator has a dimension of 80 cm \_ 25.5 cm \_ 40 cm. The fuel meat has a dimension of 80 cm \_ 14.5 cm \_ 40 cm. The total U loading is 180 Kg. The moderator for the epithermal neutron beam is made of Fluential with a thickness of 27 cm.

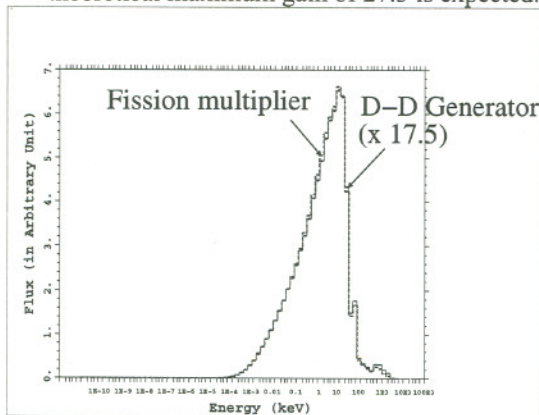
## IV. RESULTS AND DISCUSSION

The fission multipliers for the positron production system and BNCT have a  $k_{eff}$  of 0.98449 and 0.98028 respectively. Although the “true system multiplication” is  $1/(1-0.98449)=64.5$ , the absorption rate on the Cd cap of the positron production system is increased by a factor of 25 only. This number shows that the “true system multiplication” do not represent the gain in neutron flux inside the core of a fission multiplier. In fact, if light water is being irradiated with neutrons to produce heavy water, the absorption rate of neutron in light water should never be expect to increase more than  $G_f$ . When the fuel rods are removed to reduce the volume of the whole system, the flux being seen by the light water increases. This example provides a simple analogue to the positron production system. Not only that the net gain in the neutron absorption rate of the Cd cap is always less than the “true system multiplication” or the net multiplication,  $M$ , but also it is always less than the difference between  $M$  and the  $W_c$  in the fuel. The result shows that  $M$  and  $W_c$  for the fuel in the positron production system are 43.1



and 8.3 respectively. Therefore, the true theoretical maximum gain is 34.8 for the whole system.  $G_f$  for the positron production system is 35.0 so  $G_x=(M-1-G_f)=7.1$  is due to the Be(n,2n) reaction. Therefore, the theoretical maximum net gain due to the fission multiplier is  $(34.8-7.1)=27.7$  which is close 25. The remaining 10% discrepancy is due to absorption in structure material and source brightness.

The fission multiplier for BNCT has a relatively lower gain because the source brightness does not increase as much as the total neutron yield. The net gain in neutron flux at the beam port exit is  $\sim 17.5$  as shown in Figure 4.  $M$  and  $G_f$  of the D-D neutron generator driven fission multiplier for BNCT are 31.0 and 29.9 respectively. The gain from (n,xn) reaction in this system is comparatively small.  $G_x$  and  $W_c$  in the fuel are  $<0.1$  and 3.6 respectively. A theoretical maximum gain of 27.3 is expected.



**Figure 4. Epithermal neutron flux at the beam port exit of the BNCT system (Fission multiplier vs D-D neutron generator)**

The large fuel meat at the back of the neutron generator can explain why the gain in neutron brightness is smaller than its yield. The fuel meat can be separated into moderator and high-density metallic fuel assembly can be used instead of dispersion fuel. However, if the fuel is placed too close to the generator, a thicker moderator will be needed because a significant number of the fission neutrons have energy higher than 2.5 MeV. When the moderator size is increase, the geometry factor will also reduce the gain in flux at the beam port exit. If the fuel is placed far from the generator, the size of the subcritical assembly also increases and the brightness also reduces due to the geometry factor. Therefore, the fission neutrons with energy higher than 2.5 MeV are the issues in obtaining the maximum gain in this system.

## V. CONCLUSION

The FRM-II reactor described in Reference 1 has a peak thermal neutron flux of  $8 \cdot 10^{14}$  n/cm<sup>2</sup>s. The core loading for this reactor is 7.5 Kg. The subcritical multipliers have a core loading of 18.7 Kg and 180 Kg depending on the type of fuel being used. The fuel for these systems is also either highly enriched uranium or reactor-grade Pu without <sup>238</sup>U. If low-enriched uranium is used, the gain from fission is significantly reduced. A subcritical assembly fueled with U-19Pu-10Zr in the positron production system will reduce the absorption rate by half due to the reduction in source brightness. Using a light water moderator or having a low leakage design can reduce the critical mass. Unfortunately, these approaches will also significantly reduce the number of neutrons useful for the applications. The situation will not happen to a reactor because the flux in a reactor is controlled by the power. The flux from a subcritical multiplier depends on the neutron yield of the neutron generator only. In conclusion, the gain due to fission multipliers is significantly smaller than the "true system multiplication".

## ACKNOWLEDGMENT

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## REFERENCES:

- 1 C. HUGENSCHMIDT, G.KÖGEL, R. REPPER, K. SCHRECKENBACK, P. SPERR, B. STRAßER, and W. TRIFTSHÄUSER, "Monoenergetic Positron Beam at the Reactor Based Positron Source at FRM-II", *Nuclear Instruments and Methods in Physics Research B*, Vol. 192, p.97-101, Elsevier, (2002).
- 2 M. JIBALY, A. WEISS, A.R. KOYMEN, D. MEHL, L. STIBOREK, and C. LEI, "Measurement of the Positron Work Functions of the Polycrystalline Fe, Mo, Ni, Pt, Ti, and V", *Physical Review B*, Vol. 44, p. 12166, The American Physical Society, (1991).
- 3 J.M. VERBEKE, J.L. VUJIC, and K.-N. LEUNG, "Neutron Beam Optimization for

Boron Neutron Capture Therapy Using the D-D and D-T High-Energy Neutron Sources”, *Nuclear Technology*, Vol. 129, p. 257, ANS, (2000).

- 4 O.K. Harling, K.J. Riley T.H. Newton, B.A. Wilson, J.A. Bernard, L-W, Hu, E.J. Fonteneau, P.T. Menadier, E.R. Block, G.E. Koshe, Y. Ostrovsky, P.W. Stahle, P.J. Binns, and W.S. Kiger III, *The New Fission Converter Based Epithermal Neutron Irradiation Facility at MIT*, “Ninth International Symposium on Neutron Capture Therapy for Cancer”, PL-1, p. 7, Osaka, (2000).
- 5 J.F. Briesmeister, “MCNP – A General Monte Carlo N-Particle Transport Code, Version 4B”, LA-12625M, Los Alamos National Laboratory, (1997).
- 6 D. D. Keiser Jr., and M.C. Petri, “Interdiffusion Behavior in U-Pu-Zr Fuel Versus Stainless Steel Couples”, *Journal of Nuclear Materials*, Vol. 240, p. 51-61, Elsevier, (1996).
- 7 H. Koivunoro, T.P. Lou,
- 8 K.H. Kim, D.B. Lee, C.K. Kim, G.E. Hofman, and K.W. Paik, “Characterization of U-2 wt% Mo and U-10 wt% Mo Alloy Powders Prepared by Centrifugal Atomization”, *Journal of Nuclear Materials*, Vol. 245, p. 179-184, Elsevier, (1997).
- 9 K.H. Kim, J.M. Park, C.K. Kim, G.L. Hofman, and M.K. Meyer, “Irradiation Behavior of atomized U-10 wt.% Mo Alloy Aluminum Matrix Dispersion Fuel Meat at Low Temperature”, *Nuclear Engineering and Design*, Vol. 211, p. 229-235, Elsevier, (2002).