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Activation of Flibe in the HYLIFE-II
Inertial Fusion Energy Power Plant Design**

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SEQUENTIAL CHARGED-PARTICLE AND NEUTRON ACTIVATION OF FLIBE IN THE HYLIFE-II INERTIAL FUSION ENERGY POWER PLANT DESIGN

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ABSTRACT

The majority of radionuclide generation/depletion codes consider only neutron reactions and assume that charged particles, which may be generated in these reactions, deposit their energy locally without undergoing further nuclear interactions. Neglect of sequential charged-particle (x,n) reactions can lead to a large underestimation in the inventories of radionuclides which make a significant impact upon various radiological indices.¹ We have adopted the PCROSS code for use with the ACAB activation code to enable calculation of the effects of (x,n) reactions upon radionuclide inventories and inventory-related indices.^{2,3} Using this capability we have performed activation calculations for Flibe (2LiF + BeF₂) coolant in the HYLIFE-II inertial fusion energy (IFE) power plant design. For pure Flibe coolant, we find that (x,n) reactions dominate the residual contact dose rate at times of interest for maintenance and decommissioning. For impure Flibe, however, radionuclides produced directly in neutron reactions dominate the contact dose rate and (x,n) reactions do not make a significant contribution. Our results demonstrate the potential importance of (x,n) reactions and that the relative importance of (x,n) reactions varies strongly with the composition of the material considered. Future activation calculations should consider (x,n) reactions until a method for pre-determining their importance is established.

I. INTRODUCTION

The concept of (x,n) reactions was first introduced by Cierjacks and Hino.¹ These "reactions" are actually a two-step process. In the first step, a neutron interacts with an atom, and a charged particle is created. In the second step, the charged particle reacts with another atom. Often, the nuclide that is created in the second step may be produced only via complicated reaction/decay chains if only neutron reactions are considered. Figure 1 highlights the additional

daughter radionuclides which may be produced directly when (x,n) reactions are considered.

Figure 1. Isotopic daughters that require multi-step neutron reaction and decay chains may be created directly in (x,n) reactions (shaded).¹

In the HYLIFE-II and Osiris power plant designs, liquid Flibe is used as the coolant, first-wall protection scheme, and tritium-breeding material.^{4,5} Flibe is typically considered a low- to medium-activation material.^{4,7} This is because the only radiologically significant radionuclide produced from pure Flibe is ¹⁸F, which decays with a 110 minute half-life. The only long-lived radioisotope produced

in pure Flibe has been thought to be ^{10}Be . With a half-life of 1.6 million years, no energetic decay radiation, and a low production cross section, ^{10}Be does not present a significant radiological hazard. Previous studies, however, did not consider (x,n) reactions in their analyses.

Since Flibe is used for tritium breeding, many tritons and alpha particles will be generated. The alpha particles, although nearly all stop in the remaining Flibe, have an opportunity to react with fluorine via the $^{19}\text{F}(n,\alpha)^{22}\text{Na}$ reaction (2.36 MeV threshold). Since ^{22}Na has a 2.6 year half-life and emits 1.3 MeV γ -rays, the production of large quantities of ^{22}Na could significantly affect the radiological hazard posed by Flibe.

For clarity, it should be noted that ^{22}Na may also be produced in pure Flibe by neutron reactions alone via a multi-step reaction/decay chain. The dominant chain for the neutron reaction production of ^{22}Na is:

Obviously, the above reaction/decay chain is quite complicated and few ^{22}Na atoms would be produced in this manner.

II. COMPUTATIONAL METHODS

The calculational process begins with a geometric model created for use with the TART95 Monte Carlo transport code and its associated data libraries.⁸⁻¹⁰ TART95 calculates the energy-dependent neutron pathlengths in each of 175 energy groups. The neutron pathlengths are converted by the TARTREAD code into energy-dependent neutron fluences, which are used both as input to the PCROSS code and as a weighting function to collapse the 175-group neutron cross sections down into a single energy group.^{11,2} The present work uses 175-group neutron cross sections provided by the EAF3.1 cross section library.¹²

The PCROSS code permits consideration of the effects of (x,n) reactions through the use of 1-group "pseudo" cross sections.¹³ Pseudo cross sections account for the generation of charged particles in the first step and reaction of these charged particles in the second step. Pseudo cross sections are calculated by PCROSS for a particular neutron spectrum and an initial material composition. Charged-particle reactions that are currently considered by PCROSS are (p,n), (d,n), (^3He ,n), (α ,n), (d,2n), and (t,2n).¹³

PCROSS uses three different data libraries: KFKSPEC, KFKSTOP, and KFKXN. The PCROSS

development team at Karlsruhe, Germany chose to develop the KFKSPEC library due to the lack of available charged-particle spectra data.¹³ This library was generated using the ALICE code, which makes use of the pre-equilibrium hybrid model and the evaporation statistical model for compound nuclei.¹⁴ The KFKSTOP library contains stopping powers and ranges of the various charged particles and was created from an updated version of the PRAL code.¹⁵ The KFKXN library contains the charged-particle reaction cross sections and was also generated with the ALICE code.

The TARTREAD code merges the 1-group pseudo cross sections with 1-group neutron cross sections to form a combined library that is used by the ACAB radionuclide generation/depletion code.¹⁶ In addition to the radionuclide inventories, ACAB calculates the contact dose rate at the surface of a semi-infinite medium of material that contains radionuclides at the concentrations calculated for the finite quantity of material.¹⁶ Contact dose rates tend to overestimate the actual dose rates, but they are useful for comparative purposes. Figure 2 is a flowchart that shows the calculational process that has been followed in the present work.

Figure 2. In order to calculate the effects of (x,n) reactions upon radionuclide inventories, 1-group pseudo cross sections must be generated with the PCROSS code and used by the ACAB radionuclide generation/depletion code.

III. RESULTS

In order to demonstrate the potential importance of (x,n) reactions in Flibe activation, two calculations have been performed. The first calculation was performed for pure Flibe. The second calculation included Flibe's likely impurities. Both calculations utilized spectra generated for the Flibe in a one-dimensional model of the HYLIFE-II power plant design.⁴

The neutron fluxes used for both calculations were scaled by a factor of 2.15×10^{-2} , using the equivalent steady-state method, to account for the fraction of the total Flibe inventory (1240 m^3) that would be in the chamber at any given time.¹⁷ Both calculations used a 30-year operation time.

Figure 3 gives the contact dose rate as a function of time after shutdown. The figure shows that, in the case of pure Flibe, neglect of (x,n) reactions leads to severe underestimation of the contact dose rate at times after shutdown when maintenance and decommissioning would most likely be performed.

Figure 3. Without consideration of (x,n) reactions, the contact dose rate from Flibe is severely underestimated at times of interest.

The large difference in the contact dose rates with and without (x,n) reactions is due mostly to the direct production of ^{22}Na via the $^{19}\text{F}(\alpha, n)$ reaction rather than by the seven-step neutron reaction/decay chain. Despite the small value of the pseudo cross section (only $0.17 \mu\text{b}$ integrated over the neutron spectrum), ^{22}Na is much more likely to be produced directly. When (x,n) reactions are considered, 1.4×10^{14} times more ^{22}Na is produced than by neutron reactions alone. When (x,n) reactions are included, ^{22}Na reaches an equilibrium concentration of $1.3 \times 10^4 \text{ Bq/cm}^3$ (due to a large $^{22}\text{Na}(n, p)^{22}\text{Ne}$ cross section, ^{22}Na reaches equilibrium rapidly).

The second set of calculations was done for impure Flibe using a composition suggested by Toma with 10 wppm tantalum added to account for the high-Z target material that remains in the Flibe after a shot.^{18,4} Table 1 gives the Flibe composition that was used. As was done for pure Flibe, the first calculation considered only neutron reactions, and the second calculation included (x,n) reactions.

Table 1. Composition of impure Flibe.¹⁸ A full density of 2.0 g/cm^3 was assumed for the interior flows and 90% of full density was assumed for the Flibe "pocket."

Element	Weight %	Element	Weight %
Li	14.1	Be	9.1
C	0.001	N	0.001
F	76.8	Na	0.0042
S	0.0005	Cr	0.0019
Fe	0.0166	Ni	0.0026
Ta	0.0010		

For impure Flibe, the addition of (x,n) reactions makes no significant difference in the resulting contact dose rate. Figure 4 shows the contact dose rate from impure Flibe.

Figure 4. The contact dose rate from impure Flibe is dominated by activation products made in neutron reactions with impurities. For comparison, the contact dose rate from pure Flibe (including (x,n) reactions) is also shown.

Figure 5 shows the contact dose rate as a function of time after shutdown for radionuclides that make a significant contribution to the total contact dose rate from impure Flibe. Note that in impure Flibe, ^{22}Na never accounts for more than about one percent of the total contact dose rate.

Radionuclides such as ^{182}Ta and ^{60}Co dominate the dose rate at time of more than a few days after shutdown.

^{24}Na and ^{56}Mn are also larger contributors to the total contact dose rate than is ^{22}Na . The ^{182}Ta is produced from the tantalum target material that remains in the Flibe. ^{60}Co , ^{24}Na , and ^{56}Mn are produced from the nickel, sodium, and iron impurities, respectively.

Figure 5. Radionuclides produced in neutron reactions with tantalum, nickel, sodium, and iron impurities dominate the contact dose rate from impure Flibe.

IV. Conclusions

These example calculations, performed for pure and impure Flibe, show that the relative importance of (x,n) reactions upon radiological indices varies widely with the composition of materials considered. Although (x,n) reactions are of great importance for pure Flibe, they appear to be of little importance for impure Flibe. Recent publications indicate that the importance of (x,n) reactions is not limited to tritium-breeding materials.^{19,20}

These and other results suggest that future activation calculations should consider (x,n) reactions until a method is developed for pre-determining their importance in a given calculation or for a given material.

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