Adjoint Activation for Determining Source Pathways of Radioactive Inventory

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FUSION TECHNOLOGY INSTITUTE

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Abstract

Material response to neutron irradiation is a crucial aspect that must be studied in the design of a nuclear system. Neutrons can interact with various isotopes within a material and activate a stable, nonradioactive isotope into a radioactive isotope. Characteristics of the radioisotopes such as half-life, mode of decay, and decay energy will influence the activation level and safety of any nuclear design. Neutron activation is conventionally performed via a forward calculation in which the activation sequence is generated and begins with the isotopes in a material definition and then follows the pathways of activation and decay based on the activation cross sections for parent to daughter nuclear reactions. The adjoint method for activation can be performed by generating the activation sequence in the backwards direction, beginning with a daughter radioisotope and ending with the corresponding parent isotope that is at the beginning of the forward sequence. The design process can be enhanced by understanding which of the isotopes in the material definition is responsible for producing the most undesirable radioisotope. Forward activation calculations cannot directly determine the individual parent isotope percent contributions to a particular radioisotope production. In other words, it does not answer the question of how much of a certain radioisotope is produced from an isotope in the input material definition. The answer is important because impurities can be controlled or materials can then be tailored to remove the isotopes contributing the most to the production of a certain radioisotope in order to minimize its negative effect. This question can be answered using the adjoint method for performing activation analysis. This method can provide unique insight into the material selection for components in nuclear systems. This report demonstrates the use of the adjoint method for activation analysis on ARIES fusion power plant designs.

1. Background

The activation of materials from exposure to neutron environments is a significant area of study to nuclear engineering designs and safety analysis. Neutron activation is the process of converting a stable material to a radioactive material, and it is one result of neutron irradiation. It has a large influence on the waste management and operational safety of nuclear systems such as fission and fusion power plant designs. The isotopes within a given material composition will undergo nuclear reactions and radioactive decays during irradiation that alter the radioactive inventory of the material within the nuclear system. The characteristics of the radioactive inventory, such as mode of decay and decay energy, can greatly influence the design, operation, safety and cost of the system due to need for increased shielding material, remote maintenance during shutdown, or high-level waste disposal after operations. Therefore, designs of nuclear systems are often modeled using modern computational tools to accurately understand the response of materials to a neutron environment and identify the source of the undesirable radioisotopes.

Computational neutron activation analysis can be performed to model changes in isotopic composition of materials within a nuclear system. To understand the adjoint method, it is necessary to first discuss the forward method for activation analysis. A forward activation model requires a definition of the geometry, material compositions, spatial and energy neutron fluxes, irradiation schedule, and cross section data. The geometry will be defined with various regions

and each region is assigned neutron fluxes based on a certain energy group structure defined by the cross section data. The neutron fluxes are typically determined from simulations using radiation transport codes. This information is used to construct activation diagrams or trees that are used to generate the radioisotope inventory. An activation tree is a useful tool that can show reactions or decays that create unique pathways for all possible daughter isotopes and their corresponding relative concentrations that are determined from the cross section data. Each reaction or decay is considered a branch of the encompassing activation tree, and has a probability of occurring based on the cross sections or half-lives of the radioisotopes. Figure 1 illustrates an example activation tree where isotope A is activated to different radioisotopes B, C, and D that subsequently transmute or decay until a stable isotope is reached or the relative concentration of the next daughter isotope is below some threshold of interest.

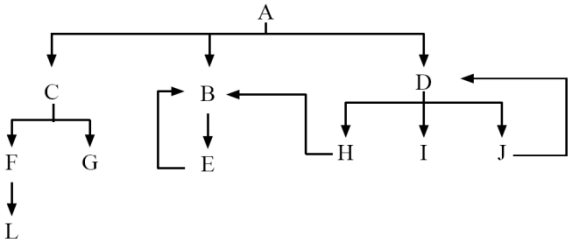


Figure 1. A sample forward activation tree [1].

The fundamental result from a forward activation calculation is the concentration of all radioisotopes resulting from irradiation in the nuclear system. Other important parameters related to activation can be determined from the radioisotope concentrations. For example, the alpha, beta, or gamma decay heat can be determined by multiplying the specific activity of a radioisotope by the average energy of decay. The decay heat is important to determine the necessary cooling requirement after shutdown to system components and maintenance to prevent biological dose hazards and radiation damage to remote handling equipment from highly radioactive components. Additionally, the radioactivity produced from a forward calculation can be compared to regulatory limits in order to determine the waste disposal rating (WDR) to indicate how the radioactive materials must be handled for waste disposal.

As mentioned earlier, the materials selection for a nuclear system will greatly influence activation results. Frequently, materials contain elements that exist either as unwanted impurities or alloying elements. Some elements may activate into a radioisotope of incredible significance even if they exist at low concentrations. Therefore, all elements must be included in the activation model to accurately determine the dominant radioisotopes resulting from activation. Often as a result of the trace elements, some activation products may result from pathways that begin at more than one of the elements in the material definition, and they will undergo distinct pathways in the activation tree. For example, stainless steels contain Mo-94 and Nb-93, both of which can activate to the radioisotope Nb-94 via (n, p) and (n, gamma) reactions, respectively.

Adjoint activation can determine the importance of various isotopes in a material definition to the production of a certain radioisotope. In the creation of the forward activation tree, the cross section data represents probabilities that a specific daughter isotope will be produced out of all of the possible daughter isotopes from a given parent isotope. Adjoint

activation is performed by creating an adjoint activation tree with relative production values of the parent isotopes rather than the daughter isotopes. To construct the adjoint activation tree, the cross section data must be inverted so that the data represents the probabilities that daughter isotopes were produced from a certain parent isotope. An example adjoint activation tree showing the reverse pathway of radioisotope L to the input isotope A is shown in Figure 2.

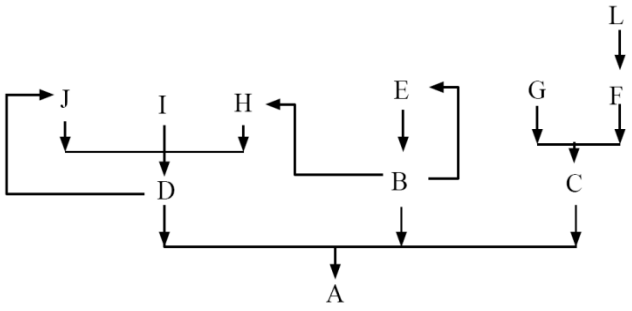


Figure 2. A sample adjoint activation tree [1].

To perform adjoint activation, a target radioisotope known to be produced in the forward calculation must be specified. Thus, it is actually a wise first step to perform the forward activation calculation prior to an adjoint calculation. However, it is not necessary if the target radioisotope is known already. This step will determine the dominant radioisotopes contributing to various parameters such as WDR and decay heat. The activation tree such as the one shown in Figure 2 is constructed for the target isotope based on the adjoint cross section data. All possible pathways of the target are created along with relative productions in terms of the various parent isotopes. It is important to note that the creation of the activation tree is independent of the material definition used in the model. There exist some pathways in the adjoint activation tree that lead to an isotope not specified in the material definition. However, the material definitions are used to determine the importance of each isotope by multiplying its number density by the relative production terms in the activation tree. For isotopes that do not exist in the material definition, the multiplication simply results in such an isotope having zero importance to the specific model. The result of the multiplication is the amount of the parent isotope that contributes to the production of the daughter radioisotope.

The background regarding the derivation of the adjoint operator for activation has been developed previously by White [2]. The implications of the adjoint activation are that optimal material compositions can be easily determined to minimize safety and environmental impact. For example, the isotopic concentrations in a material can be determined to ensure a WDR that indicates low-level waste disposal or identify decay heat trends that allow maintenance to be safely performed quickly after shutdown to avoid long cooling periods.

2. Methodology and Codes

Models of ARIES-ACT and ARIES-CS power plants [3] are used to perform adjoint activation. The ARIES-ACT divertor [4] is used for adjoint WDR analysis, and the ARIES-CS LiPb blanket [5] is used to perform adjoint decay heat analysis for alpha, beta, and gamma decay.

The ARIES-ACT divertor is taken as a 1-D cylindrical model used in this analysis. It consists of 14 MeV neutrons from a plasma impinging on the divertor radial build made of 0.5 cm W armor and 7.2 cm cooling channel (refer to Fig. 3). This design averages a neutron wall loading (NWL) of 1 MW/m^2 over the divertor surface. The operation pulse schedule is modeled for 85% availability and assigns a lifetime to the divertor of 3.4 full power years (FPY), meaning 4 years of operation before the divertor must be replaced.

The W in both armor and cooling channel is composed of W with nominal impurities shown in Table 1. Included in the nominal impurities of W was Nb. Knowing that the Nb content in W creates activation problems, calculations were made using both 1 and 5 wppm Nb in the W of the armor and cooling channel. The WDRs were tabulated at 100 years after shutdown.

The ARIES-CS blanket is modeled as a 1-D cylindrical model with a 14 MeV plasma neutron source. The complete radial build is shown in Figure 4. In this analysis, the blanket experiences the same operation schedule as the divertor wherein it is designed with a lifetime of 3.4 FPY and an average NWL of 2.6 MW/m^2. The 63 cm blanket is composed of the 3.8 cm first wall (FW), 54.2 cm of homogenized breeder region, and 5 cm back wall (BW). The breeder region is composed of the LiPb breeder, SiC flow channel inserts, and ferritic steel cooling channels. The FW and BW of the blanket are used in the adjoint analysis. The material compositions of these layers with a complete list of alloying elements and impurities are shown in Table 2. The FW and BW decay heat is examined at one day after shutdown.

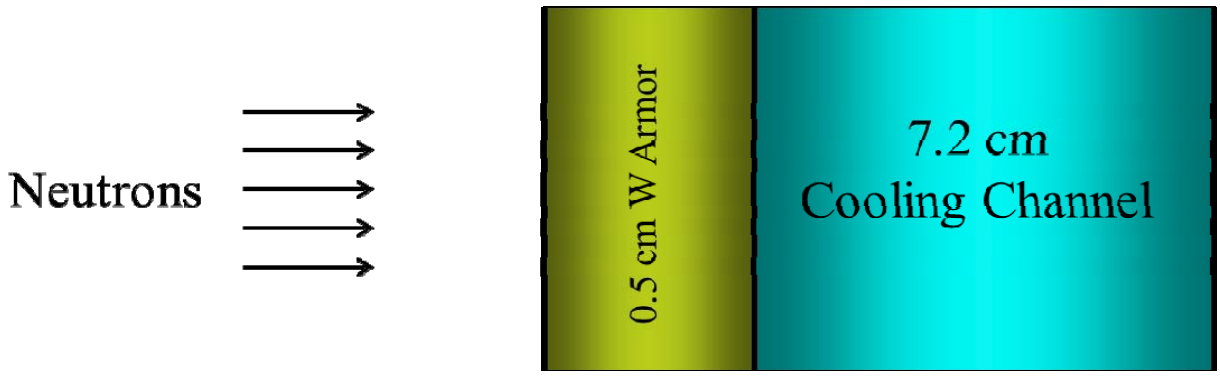


Figure 3. Radial build of divertor to be used in WDR calculations [4, 6].

Table 1. ARIES-ACT divertor composition and W impurities. Analysis with Nb impurity content of 5 wppm and 1 wppm to demonstrate the shift in dominant isotopes [6].

Divertor Composition (by volume):

W armor	88% W 12% void
Cooling Channel	32.2% W 11.6% FS 56.2% He coolant

W Impurities (in wppm)

C	10	Ba	<2	Na	<2
H	2	Ca	<2	Nb	1 or 5*
N	<2	Cd	<2	Mo	20
O	5	Co	<2	Ni	<2
P	<10	Cr	<2	Pb	<2
S	<2	Cu	<2	Ta	<10
Si	5	Fe	10	Ti	<2
Ag	<5	K	5	Zn	<2
Al	5	Mg	<2	Zr	<2
As	<2	Mn	<2		

*Calculations performed using both 1 and 5 wppm Nb

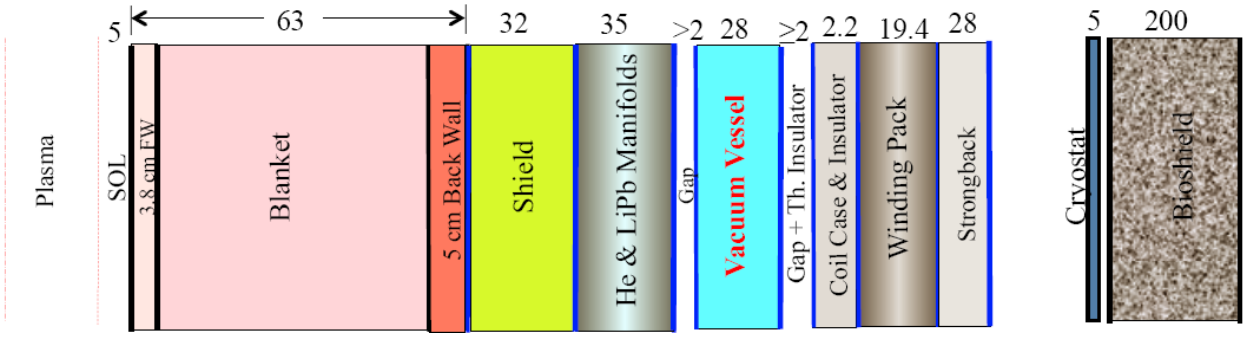


Figure 4. Radial build of blanket to be used in decay heat calculations. The FW and BW of the blanket region are used in the adjoint calculation [4].

A coupling of two computer codes was used to simulate the neutron transport and activation analyses. The neutron transport was performed by PARTISN, a discrete ordinate, neutral particle transport code [7]. PARTISN modeled the system using the $S_{12}P_5$ approximation along with a 1-D cylindrical equivalent of the ARIES divertor and blanket models illustrated in Figures 2 and 3. The forward and adjoint activation analysis was handled by ALARA [1, 8]. The FENDL-2 nuclear data and activation libraries consisting of 175 neutron and 42 gamma groups

Table 2. ARIES-CS Blanket FW and BW composition.

Blanket (by volume)							
First Wall (FW)		8% ODS-MF82H 26% MF82H 66% He					
Back Wall (BW)		80% MF82H 20% He					
MF82H (by weight) **7.89 g/cm^3**				**ODS-MF82H (by weight)** **7.78 g/cm^3**			
C	0.1	Cd	4.00E-05	B	3.00E-04	Nb	3.30E-04
Al	1.40E-03	Ta	0.02	C	0.04	Mo	2.10E-03
V	0.2	W	2	N	5.00E-03	Pd	5.00E-06
Cr	7.5	Os	5.00E-06	O	0.13	Ag	1.00E-05
Fe	90.11586	Ir	5.00E-06	Al	0.01	Cd	4.00E-05
Co	2.80E-03	Bi	2.00E-05	Si	0.24	Sn	1.00E-03
Ni	4.74E-02	Eu	5.00E-06	P	5.00E-03	Sb	5.00E-04
Cu	1.00E-02	Tb	2.00E-06	S	2.00E-03	Ta	0.08
Nb	3.30E-04	Dy	5.00E-06	Ti	0.09	W	2
Mo	2.10E-03	Ho	5.00E-06	V	0.29	Os	5.00E-06
Pd	5.00E-06	Er	5.00E-06	Cr	8.7	Ir	5.00E-06
Ag	1.00E-05	U	5.00E-06	Mn	0.45	Bi	2.00E-05
				Fe	87.891458	Eu	5.00E-06
				Co	2.80E-03	Tb	2.00E-06
				Ni	4.74E-02	Dy	5.00E-06
				Cu	0.01	Ho	5.00E-06
				As	2.00E-03	Er	5.00E-06
				Y	0.7	U	5.00E-06

were used [9, 10]. ALARA provided the capability of modeling the actual operating schedule of 85% yearly availability of the machine over the course of 4 years – the lifetime of the replaceable divertor and blanket. More importantly, ALARA was capable of performing the adjoint activation calculations pertinent to this work. An appendix is provided at the end of this report to offer guidance on converting a forward ALARA input file to an adjoint ALARA input file.

3. Results

Adjoint activation was performed to characterize the WDR and decay heat resulting from alpha, beta, and gamma decay. As mentioned earlier, the ARIES-ACT divertor was used as the model to examine the WDR and the blanket was used to examine the various decay heats. The following sections document the results from this analysis.

3.1. WDR Results

The ARIES-ACT divertor was used to perform adjoint activation. It has a WDR of 0.98 and qualifies as Class C low-level waste. The forward mode was previously performed and those results were used to determine the dominant radioisotopes that contributed to the WDR [6]. The dominant isotopes in the W-armor and cooling channel were Nb-94, Ag-108m, Tc-99, and Re-186m. Each of these radioisotopes was used in a separate calculation as the target isotope for the adjoint activation calculation. Tables 3 and 4 show the results of this analysis for 5 and 1 wppm Nb impurity, respectively. Shown in these tables are the percent contributions of the two dominant parent isotopes in the material definition to the production of the target, as is unique to the adjoint method analysis. For instance, Nb-93 is responsible for 97% of the Nb-94 produced in the activation of the W-armor given 5 wppm as shown in Table 3. Similarly, Mo-94 is responsible for 2.3% of the Nb-94. Furthermore, the percent contribution of each target radioisotope to the total WDR of each component is shown. The percent contribution results in Table 3 for the cooling channel correspond to the values determined previously given in Table 2 of Ref. [11].

Table 3. WDR adjoint activation results for ARIES-ACT divertor given 5 wppm Nb impurity at 100 years after shutdown.

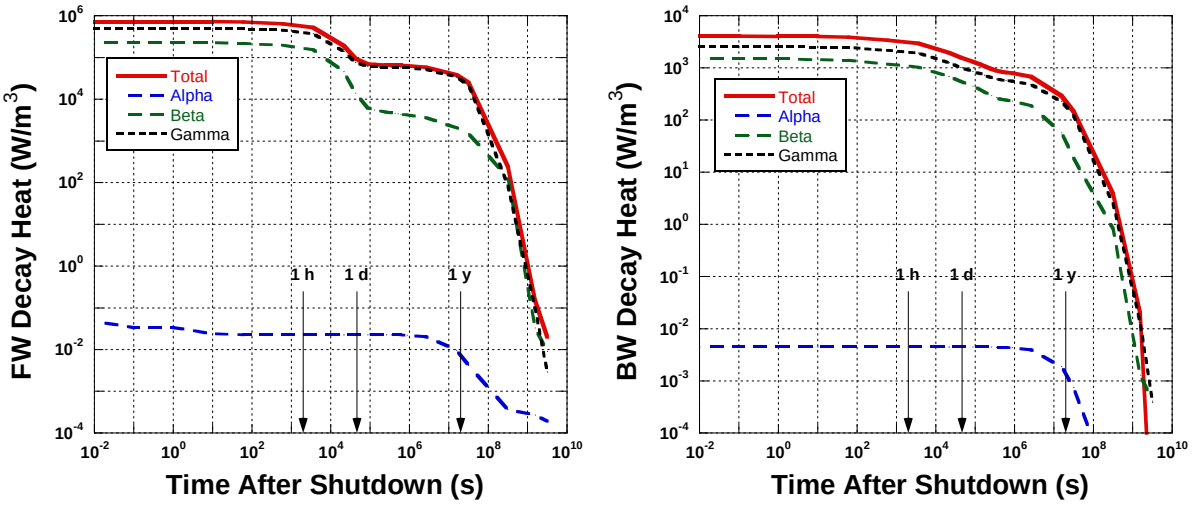
	W-armor		Cooling Channel	
Target	Parent (%)	% contribution to total	Parent (%)	% contribution to total
Nb-94	Nb-93 (97.0%) Mo-94 (2.3%)	42.9%	Nb-93 (98.1%) Mo-94 (1.5%)	53.9%
Ag-108m	Ag-109 (94.1%) Ag-107 (5.9%)	39.5%	Ag-109 (90.8%) Ag-107 (9.2%)	28.4%
Tc-99	Mo-100 (57.3%) Mo-98 (42.6%)	14.3%	Mo-98 (54.9%) Mo-100 (44.9%)	14.6%
Re-186m	W-186 (95.5%) W-184 (4.5%)	2.3%	W-186 (93.2%) W-184 (6.8%)	1.6%

Table 4. WDR adjoint activation results for ARIES-ACT divertor given 1 wppm Nb impurity at 100 years after shutdown.

	W-armor		Cooling Channel	
Target	Parent (%)	% contribution to total	Parent (%)	% contribution to total
Ag-108m	Ag-109 (94.1%) Ag-107 (5.9%)	59.2%	Ag-109 (90.8%) Ag-107 (9.2%)	46.3%
Tc-99	Mo-100 (57.3%) Mo-98 (42.6%)	21.4%	Nb-93 (93.5%) Mo-94 (5.2%)	24.9%
Nb-94	Nb-93 (86.7%) Mo-94 (10.3%)	14.4%	Mo-98 (54.9%) Mo-100 (44.9%)	23.8%
Re-186m	W-186 (95.5%) W-184 (4.5%)	3.4%	W-186 (93.2%) W-184 (6.8%)	2.6%

3.2. Decay Heat Results

Adjoint activation was performed for the FW and BW of the ARIES-CS blanket. Figure 5 illustrates the decay heat trends for various cooling periods for the FW and BW. The decay heat is most important at one day after shutdown because that is the desired cooling time before maintenance should begin.



(a) (b)

Figure 5. (a) ARIES-CS FW decay heat results. (b) ARIES-CS BW decay heat results.

3.2.1 Gamma Decay Heating Results

The gamma decay heating is the largest contributor to the total decay heat in the blanket. Figure 6 shows the trend of the gamma heating ranging from shutdown to 100 years after shutdown. Only the two most dominant isotopes are recorded for each target isotope. The dominant radioisotopes contributing to the gamma heating were determined to be Mn-54, Ta-182, and Cr-51 in the FW. Similarly, W-187, Mn-54, Fe-59 contributed most to the gamma decay heat in the BW. Each of these radioisotopes undergoes gamma decay, which contribute to the total gamma decay heat.

The results from the adjoint activation indicate the percent contribution of the parent isotopes that are responsible for the dominant radioisotopes undergoing gamma decay. The complete results are shown in Tables 5 and 6. For example, Mn-54 is responsible for 77.2% of the gamma decay heat in the FW, and it is produced via an (n,p) reaction with Fe-54. The isotope Fe-54 is responsible for 92.1% of Mn-54 that is produced. Furthermore, W-186 and Fe-54 activate into the radioisotopes mentioned previously that contribute the most to the gamma decay heating of the BW. The isotope W-186 transmutes to W-187 via an (n,gamma) reaction.

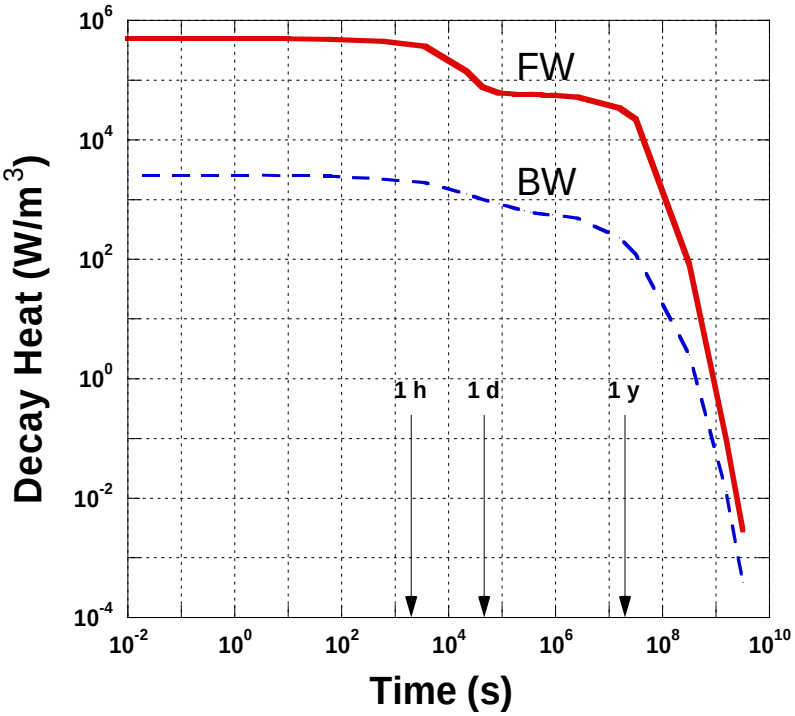


Figure 6. Gamma decay heat in the FW and BW of the ARIES-CS blanket.

Table 5. FW gamma decay heat results from adjoint activation at 1 day after shutdown.

FW Gamma Heating		
Target	Parent (%)	% contribution to total
Mn-54	Fe-54 (92.1%) Fe-56 (5.1%)	77.2%
Ta-182	Ta-181 (67.6%) W-182 (32.0%)	3.1%
Cr-51	Cr-52 (76.3%) Fe-54 (19.5%)	2.9%

Table 6. BW gamma decay heat results from adjoint activation at 1 day after shutdown.

BW Gamma Heating		
Target	Parent (%)	% contribution to total
W-187	W-186 (100%)	30.2%
Mn-54	Fe-54 (100%)	22.5%
Fe-59	Fe-58 (85.1%) Fe-57 (14.8%)	16.1%

3.2.2 Beta Decay Heating Results

The beta heating has a slightly less significant contribution than the gamma decay heating in both the FW and BW of the ARIES-CS blanket. Figure 7 shows the trend of the beta heating ranging from shutdown to 100 years after shutdown.

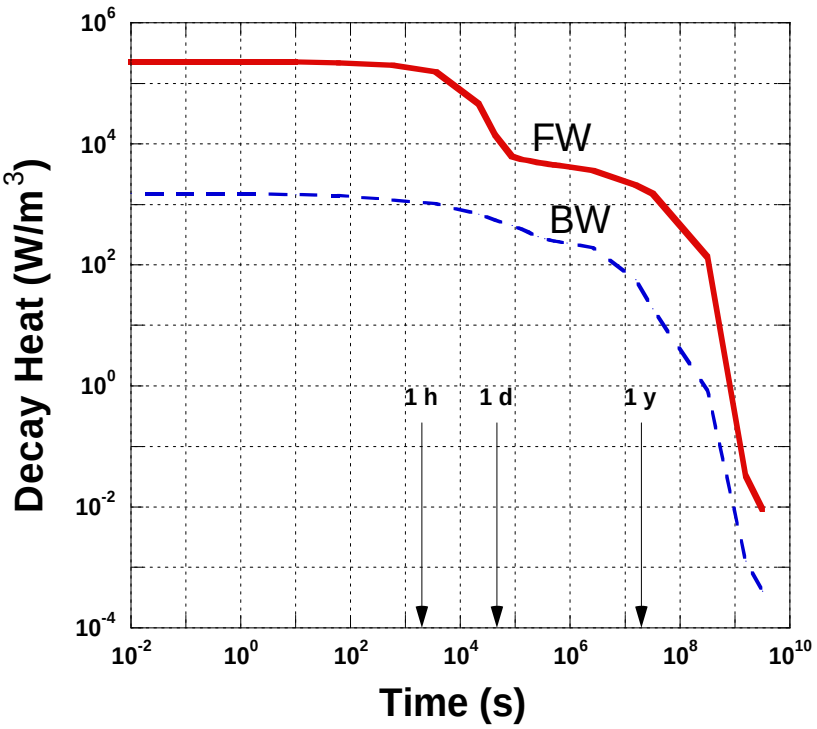


Figure 7. Beta decay heat in the FW and BW of the ARIES-CS blanket.

From the forward calculation, the dominant radioisotopes contributing to the beta decay heating were determined to be Fe-55, W-185, W-187, and Ta-182 in both the FW and BW. Each of the most dominant radioisotopes was used as a target for the adjoint activation calculations. The results indicate that the most dominant radioisotope, Fe-55, that undergoes beta decay in the FW is transmuted from the Fe-56 and Fe-54 via an (n, 2n) and (n, gamma) reactions, respectively. Additionally, the W-184 and W-186 in the material definition are most responsible for the production of W-185 and W-187. The complete results are shown in Tables 7 and 8.

Table 7. FW beta decay heat results from adjoint activation at 1 day after shutdown.

FW Beta Heating		
Target	Parent (%)	% contribution to total
Fe-55	Fe-56 (98.0%) Fe-54 (1.9%)	26.7%
W-185	W-186 (99.6) W-184 (0.4%)	25.9%
W-187	W-186 (100%)	10.2%

Table 8. BW beta decay heat results from adjoint activation at 1 day after shutdown.

BW Beta Heating		
Target	Parent (%)	% contribution to total
W-185	W-184 (97.9%) W-186 (1.4%)	40.4%
W-187	W-186 (100%)	39.5%
Ta-182	Ta-181 (99.7%) W-182 (0.2%)	8.6%

3.2.3 Alpha Decay Heating Results

Figure 8 shows the trend of the alpha heating ranging from shutdown to 100 years. From the forward calculation, the dominant radioisotopes contributing to the alpha heating at one day after shutdown were determined to be Po-210 and Pu-238. The total alpha heating is insignificant to the total decay heat, but it is included here to demonstrate the adjoint activation method.

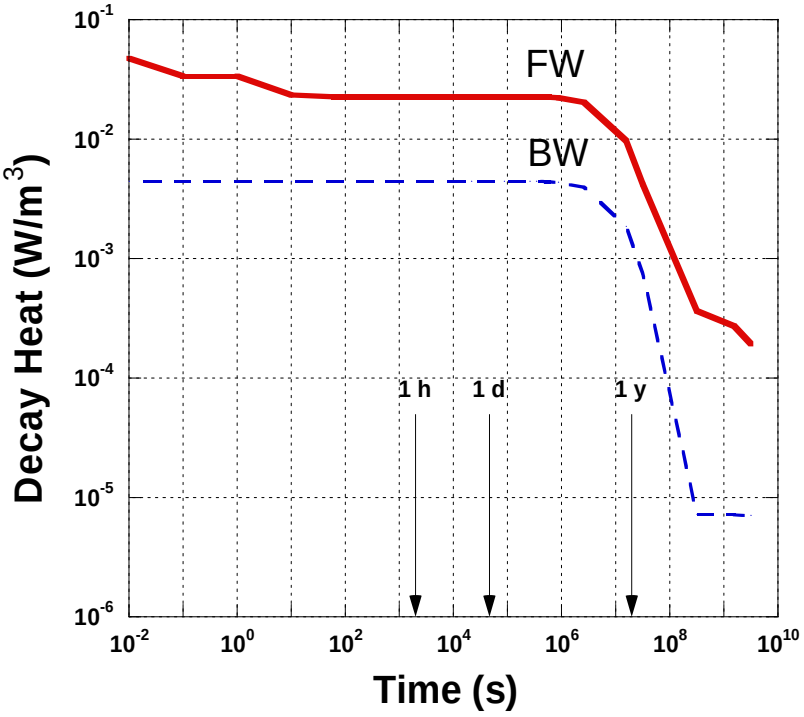


Figure 8. Alpha decay heat in the FW and BW of the ARIES-CS blanket.

Each of the most dominant radioisotopes was used as a target for the adjoint activation in the FW and BW. The results indicate that the most dominant radioisotope, Po-210, undergoing alpha decay in both the FW and BW, is produced from an (n, gamma) reaction with Bi-209 followed by a beta decay of Bi-210. The results are shown in Tables 9 and 10.

Table 9. FW alpha decay heat results from adjoint activation at 1 day after shutdown.

FW Alpha Heating		
Target	Parent (%)	% contribution to total
Po-210	Bi-209 (100%)	98.0%
Pu-238	U-238 (99.9%) U-235 (0.1%)	1.60%

Table 10. BW alpha decay heat results from adjoint activation at 1 day after shutdown.

BW Alpha Heating		
Target	Parent (%)	% contribution to total
Po-210	Bi-209 (100%)	98.8%
Pu-239	U-238 (100%)	0.15%

4. Conclusions

Neutron activation analysis is a critical step in determining the safety parameters and waste disposal options for nuclear systems. The adjoint method provides unique insight to the response of materials to irradiation by determining the isotopes in a material definition that contribute to the production of a given radioisotope. With the use of adjoint activation, appropriately tailored materials can be determined to increase the margin of safety of important design parameters such as WDR and decay heat. The ARIES power plant designs are examined in this report to demonstrate the use of the adjoint method for activation. Future work could examine effects on source pathways of radioactive inventories from combining components. It is clear that the adjoint method for activation is a powerful tool in design and analysis of material compositions for nuclear systems.

Appendix

This appendix provides instructions regarding how to convert an ALARA forward input problem to an adjoint problem. As mentioned, it is essential to perform a forward calculation first if the dominant radioisotopes for a given output parameter are not known already. There are a few lines in the ALARA input file that must be added or changed in order to indicate an adjoint problem.

1. The “data_library” definition must be modified to indicate an adjoint data library. For example, the definition could be changed to the following,

```
data_library     adjlib     fendl2adj
```

   where the second keyword indicates adjoint library treatment and the last keyword is the name of the adjoint library located within the working directory.

2. The second addition is within the mixture definition of the zone in which you wish to perform adjoint analysis. The target isotope indicator that goes within a given mixture definition must be specified. Only one mixture can be solved for at a time. For example, if a zone contains the mixture “w-armr” that is known to produce Nb-94, then the mixture definition to perform adjoint analysis would appear like the following,

```
mixture    w-armr
     material  W_Impurities  1.0  .884
     target isotope nb-94
end
```

It is required in the input file to solve for only the zone that contains the mixture the user has placed the “target” indicator. It is possible to solve for more than one zone if the zones contain the same mixture. After these changes are made, ALARA can be executed from the command line in the same fashion as that of a forward calculation.

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