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RADIOACTIVITY AND NEUTRON DAMAGE CALCULATORS FOR RESEARCH AND TEST REACTOR NEUTRON-ACTIVATED MATERIALS

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The Idaho National Laboratory Nuclear Science User Facilities program is proposing to launch a new online expert system that will assist users in planning experiments and writing proposals. The Combined Materials Experimentation Toolkit (CoMET) will include two nuclear calculators. The first of these, the Neutron Damage Calculator, will assist users in selecting reactor positions in order to obtain desired material damage in Damage Per Atom (DPA) and provide the required irradiation time to reach the damage levels. The second, the Radioactivity Calculator, will estimate the post-irradiation radioactivity levels of sample materials, which will inform technical staff of examination facility requirements and any potential scheduling delays. Both of these calculators were designed to simplify the information needed from users in order to properly plan and execute awarded proposals. This paper presents the primary purpose and methodologies implemented by these two calculators.

I. INTRODUCTION

The Idaho National Laboratory (INL) Nuclear Science User Facilities (NSUF) program supports nuclear energy-relevant research into the effects of radiation on nuclear fuels and structural materials. The NSUF provides access, at no cost to the researcher, to the Advanced Test Reactor (ATR), the High-Flux Isotope Reactor (HFIR), and eight other university and national laboratory research and test reactors for radiation effects experiments. Proposals to the NSUF include desired experimental conditions, including neutron flux, material damage and/or temperature, and specify any post-irradiation examination (PIE) to be performed. The execution of an awarded proposal is a complex undertaking involving the coordination of NSUF technical staff with scientists and operators at partner facilities and with the principal investigators. A full project, involving neutron irradiation and subsequent PIE, can cost up to four million dollars (US) and run for seven years. It is unreasonable to expect that a sole investigator would be able to include all of the necessary information in their proposal without guidance. In order to optimize and assist researchers with the proposal writing and experimental design processes, the NSUF is creating a set of online tools called the CoMET—the Combined Materials Experimentation Toolkit.

CoMET includes four databases to support nuclear energy research. First is the Nuclear Energy Infrastructure Database (NEID), which contains capability data and contact information for 500 nuclear fuels and materials R&D facilities, both within the U.S. and internationally. The second database is the Nuclear Fuels and Materials Library (NFML), which has data on approximately 10,000 specimens that were previously irradiated in NSUF projects, legacy reactor projects (such as EBR-II), or commercial specimens. The NFML also includes the actual specimens that are available for use in NSUF projects. Two other databases are currently under construction, a collaboration database with information on NSUF researchers and scientific staff at the NSUF facilities and a research projects database that includes previously completed NSUF projects. An expert system is being implemented to tie these resources together and aid researchers in designing, planning, writing and submitting strong research proposals to one of NSUFs competitive proposal solicitations. All of these tools will be available on the NSUF website (nsuf.inl.gov).

One of the greatest disconnects encountered when executing an awarded proposal is between the material scientist submitting it and the nuclear engineers designing the irradiation experiments and examinations procedures. This disconnect is due to different terminology usages and required or desired information. Part of the CoMET online expert system is a tool for assisting the researcher in selecting the appropriate reactor and reactor position in order to obtain the desired material damage or neutron flux levels. This tool, the Neutron Damage Calculator, will facilitate communication and understanding between the researcher and design engineering by providing outputs in units familiar to each party. Another benefit of the Neutron Damage Calculator is its ability to provide researchers with several reactor position options, which could attain similar flux and/or desired damage levels. This benefit lessens the demand for ATR experimental positions, which have long wait times and high costs, and increases awareness of other reactor positions that can accomplish the desired effects.

Another component of the CoMET system estimates the post-irradiation radioactivity of the sample material. This tool is used to inform researchers and technical leads of potential scheduling and safety issues resulting from the radioactivity levels of the sample. Using the output provided by the Radioactivity Calculator, PIE activities may be scheduled at the appropriate facility which will have the capabilities to handle materials of the estimated radioactivity level. The ability to estimate these parameters at the beginning of the scoping and planning process can be expected to save time, money, and personnel radiation dosages.

This paper will specifically discuss the purpose and methodology of the two tools mentioned above—the Neutron Damage Calculator and the Radioactivity Calculator.

II. NEUTRON DAMAGE CALCULATOR

II.A. Purpose

As discussed in the introduction, users have access to multiple NSUF research reactors for material irradiation experimentation. When applying for assistance from the NSUF, users can propose the experiment, including time frame, material to be irradiated and/or where they wish to perform the experiment. Within the available research reactors there are hundreds of possible irradiation positions. These positions all have different neutron flux conditions that will affect the experiment. The Neutron Damage Calculator (NDC) was constructed to assist users in determining the optimal reactor and reactor position for the desired material effects. Currently, the neutron flux and damage calculations are being applied to ATR and HFIR. As a baseline measurement of material damage, this tool uses displacements per atom (DPA) to inform the user of the probable amount of damage incurred by a material, while indicating the best possible location and run time for their proposed experiment. The NDC will be part of the CoMET online expert system and will allow users to input irradiation time, location within the reactor or desired DPA.

II.B. Method

The calculator was developed during the summer of 2017 at INL. For DPA calculations, 41 isotopes displacement damage-energy cross-sections were calculated using SPECTER. SPECTER was developed by Lawrence R. Greenwood and Robert K. Smither, and is designed to calculate "spectral averaged displacements, recoil energy, gas production, and total damage energy."[1] The SPECTER manual contains the 41 isotopes and cross-sections used by the code. The NDC uses these cross-sections as the primary source for displacement cross-section calculations, which are needed to calculate DPA. SPECTER can calculate DPA from a spectrum of neutron energies.[1] However, development of a separate DPA calculator was decided upon in order to create a quick and easy way for users to understand the achievable DPA. While SPECTER is a tried and true method for calculating material damage, it does have limitations in run time, the need to install and learn a new program, and complex output files.

Equation 1 shows the DPA rate, which is dependent on the energy of the incident neutrons E_i and the total neutron flux Φ.[2,3] The displacement cross-section σ_D, due to its dependence on the energy spectrum and the material, must be obtained from SPECTER and is based on the energy transferred from an incoming neutron and the target nucleus, the energy lost during the interaction and the actual displacements created.[4] Therefore, the DPA rate R_{dpa} is found by integrating over the incident neutron energies (E_m to E_M) and is expressed as

$$R_{dpa} = \int_{E_m}^{E_M} \sigma_D(E_i)\Phi(E_i)\, dE_i, \tag{1}$$

and can be approximated as[2]

$$R_{dpa} = \sum_{i=1}^{N} \sigma_{D_i}\phi_i. \tag{2}$$

While there are several methods used to calculate the variables in the DPA equation, the most commonly used method is the Norgett, Robinson, Torrens (NRT) model. The NRT method, utilized by SPECTER and the NDC, is based on the Kinchin-Pease (K-P) model[5]

$$N_d = \left\{ \begin{array}{cc} 0 & 0 < E < E_d \\ 1 & E_d < E < 2E_d \\ \frac{E}{2E_d} & 2E_d < E < 1 \\ \frac{E_i}{2E_d} & E_i < E < \infty \end{array} \right\}. \tag{3}$$

The K-P model is commonly used to calculate the number of displaced atoms N_d caused by an interaction.[6] In Equation 3, E is the energy of the primary knock on atom (PKA), E_d is the threshold damage energy and E_i is the energy above which the atom is only slowed from electron excitation. The NRT model simplifies the K-P methods as follows[6]

$$N_d = \frac{\kappa \hat{E}}{2E_d}, \tag{4}$$

where κ is the displacement efficiency and is set to 0.8 for the NRT method. $\hat{E}$ is the energy available from the elastic collisions to cause displacements and is calculated using the following NRT equations

$$a = \left(\frac{9\pi^2}{128}\right)^{1/3} a_0 \left(Z_1^{2/3} + Z_2^{2/3}\right)^{-1/2} \tag{5}$$

$$\epsilon = \left[\frac{A_2 E}{A_1 + A_2}\right]\left[\frac{a}{Z_1 Z_2 e^2}\right] \tag{6}$$

$$g(\epsilon) = 3.4008\epsilon^{1/6} + 0.40244\epsilon^{3/4} + \epsilon \tag{7}$$

$$k = 0.1337 * Z^{1/6}\left(\frac{Z_1}{A_1}\right)^{1/2} \tag{8}$$

$$\hat{E} = \frac{E}{[1 + kg(\epsilon)]}. \tag{9}$$

Equations 5–8, (a, ϵ, $g(\epsilon)$ and k) are intermediate steps required to find the energy $\hat{E}$ available from the elastic collision in Equation 9. Within these equations, a_0 is the Bohr radius, Z is the atomic number, A is the atomic mass of the neutron or the target atom and e is the charge.[6] These equations illustrate all of the information considered when calculating the displacement cross-sections required to estimate the DPA. While there are many other factors contributing to the displacement calculation, further derivation is outside the scope of this paper.

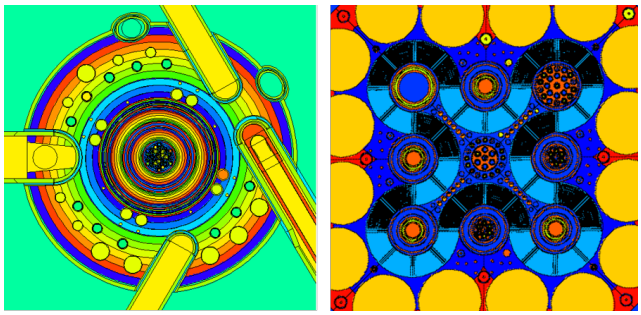

Fig. 1. Full core HFIR and ATR MCNP models

The NDC utilizes these scientific methods in an indirect way. SPECTER uses the NRT method to calculate the damage and the displacement cross-sections and displacement damage-energy cross-sections, whereas the NDC uses the calculated cross-sections to calculate DPA.[6] Taking the displacement damage-energy cross-sections, the calculator only needs to multiply by $0.8/2E_d$, as in the NRT method, to output the displacement cross-section needed for the DPA calculation.

II.C. MCNP Models and Applied Reactor Positions

The other factor needed to calculate DPA rate is the neutron flux. Neutron flux, or simply flux, was calculated using the general-purpose Monte-Carlo modeling and simulation code, MCNP. Using the F4 tally, the NDC uses the flux output to calculate DPA. Provided below is the necessary multiplier to convert the particles/cm^2 produced by MCNP to the required flux value in neutrons/cm^2/s.[7]

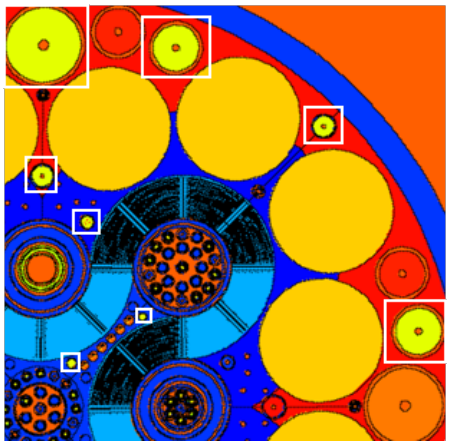

Fig. 2. Selected experimental positions in the INL ATR core

$$\text{Flux} = \text{F4 output} * \frac{2.43 \text{ fission neutrons}}{\text{fission}} * \frac{1 \text{ fission}}{200 \text{ MeV}} * \frac{1 \text{ MeV}}{1.60219x10^{-13} \text{ J}} * \frac{1 \text{ J}}{1 \text{ W-s}} * \text{Power} \quad (10)$$

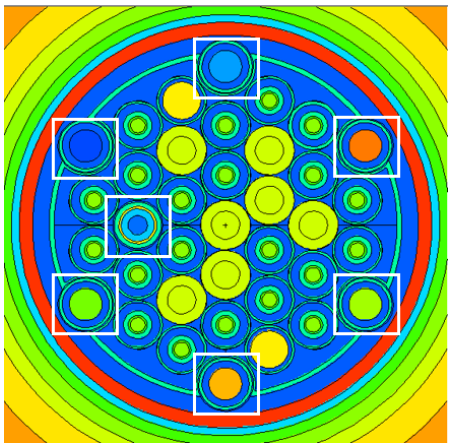

Fig. 3. Selected experimental positions in the HFIR center flux trap

Users are not required to have or run an MCNP deck for each reactor. The flux calculations were completed in advance in order to simplify calculator usage by outputting the flux of each position and material to a library referenced by the NDC.

As mentioned previously, the two current reactors in the tool are ATR and HFIR. The reactor positions available to users are highlighted (outlined with white squares) in Figures 1–4. Positions in ATR are in the northeast corner of the reactor. HFIR positions are representative of symmetric positions within the reactor where the flux and DPA are assumed to be comparable.

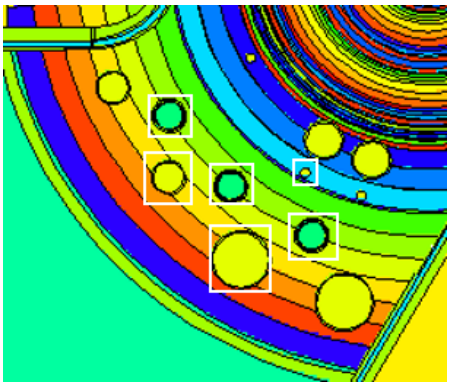

Fig. 4. Selected experimental positions in the HFIR outer-core positions

II.D. Results

User input is simple and straightforward; however, the user must first decide on the desired output: irradiation time, reactor position or the DPA. As shown Figure 5, the user first selects the desired output, then follow-up questions are posed to further refine the option selected. After receiving the required input, the NDC will perform the DPA and/or time calculations and return the desired results.

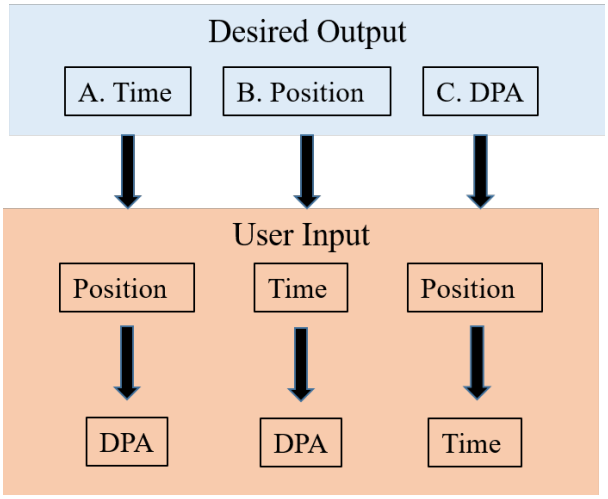

Fig. 5. Calculation flow of the NDC based on user input

Figure 6 shows the current calculator interface and provides an example of the required input and follow-up questions asked based on a situation where the user wanted to know how many days it would take to reach a certain DPA in a specific position of ATR.

```
WHAT DO YOU NEED TO KNOW FROM THE DPA CALCULATOR?
A. TIME
B. EXPERIMENTAL POSITION
C. DPA

Chosen otion:A
What is the desired material?Fe
Please select from positions:,1A,1B,9B,13A,5I,3I,1I,21I
Position:1B
Apporximately, what DPA do you want to reach?0.5
It would take [256.13335905] days to reach this DPA in position1B.
>>>
```

Fig. 6. Example of user input for the NDC

II.E. Validation

Displacement cross-section data from two other sources were collected and compared to the output from SPECTER to verify the DPA results. One source is the displacement cross-section from the Evaluated Nuclear Data File (ENDF) libraries maintained by the National Nuclear Data Center (NNDC) and the International Atomic Energy Agency (IAEA).[8] The other values were calculated using NJOY2016's HEATR module. HEATR calculates the available energy from a reaction needed to create displacements and outputs the damage energy production cross-section in eV-barns.[9] When multiplying the available energy output from HEATR with the $0.8/2E_d$ from the NRT method, the output is the displacement cross-section.[10] Figure 7 shows the relationship between the displacement cross-sections of SPECTER, NJOY, and the ENDF library for iron. Comparisons for all 41 isotopes were performed; however, only iron is presented here for brevity.

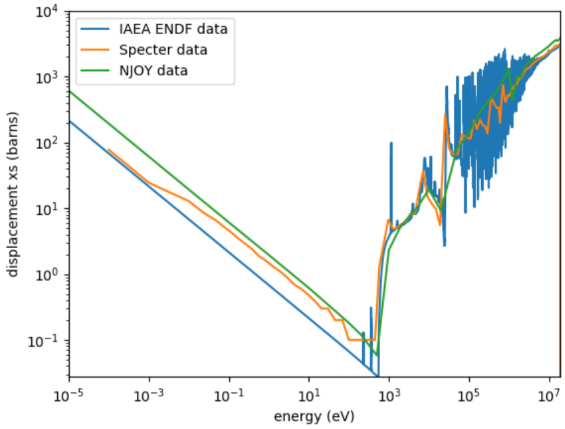

Fig. 7. Comparisons of NJOY, ENDF, and SPECTER cross-section data for iron

As shown in Figure 7, the graphical trend of the displacement cross-sections are comparable between the three data sources. The main discrepancy lies in the thermal neutron region. While this is not true for all isotopes, it is a factor that will be explored in future validation of the NDC. However, the data comparison does illustrate an important point:

the results of the DPA calculation will vary depending on the cross-section data used. Users need to be aware of this factor when utilizing the tool.

II.F. Impact

The goal of the NDC was to create a tool for proposal writers and NSUF facility users that would provide a baseline understanding of the possible material damage associated with available reactor positions. Use of the NDC will inform researchers of the best reactor position options for their proposals and provide NSUF technical staff with approximate neutron flux and damage values. By incorporating the NDC into the online CoMET system, the barrier of installing and learning a new program is removed, and users can more easily obtain DPA estimates.

III. RADIOACTIVITY CALCULATOR

III.A. Purpose

Through the NSUF and its associated user facilities, researchers also have access to a variety of hot cells and material characterization equipment at national laboratories, universities and industrial sites across the United States. In order to estimate the post-irradiation radioactivity levels of materials early in the experiment cycle, the Radioactivity Calculator was created. Post-irradiation examination (PIE) of materials poses some difficulties, specifically when studying neutron-irradiated materials. Inside a test reactor, under high neutron flux, stable materials can activate (capture a neutron) and become unstable or radioactive. These newly created radioactive materials then decay, based on their respective half-lives, and emit energetic radiation. Nuclides with short half-lives are able to decay to sufficiently low radiation levels prior to the sample being removed from the reactor. However, nuclides with longer half-lives, such as cobalt-60, present a challenge since they must be handled with extreme caution or stored until their radioactivity levels decrease. If immediate examination is required, the examination must take place in a hot cell, which can dramatically increase the cost of the project. On the other hand, storage can greatly increase the project time-line. In order to obtain an estimate of the post-irradiation radioactivity levels of material samples, that will inform researchers and technical leads of potential safety and/or scheduling issues, the Radioactivity Calculator was created based on three activation and decay stages.

III.B. Method

As of May 2018, the Radioactivity Calculator contains data for 92 elements and 760 nuclides. Elements currently not represented in the calculator include H, He, Li, At, Cf, Es, Fm, Md, No, Lr, Rf, Db, Sg, Bh, Hs, Mt, Ds, Rg, Cn, Uut, Fl, Uup, Lv, Uus and Uuo. For all included nuclides, a database with nuclide atomic weight (g/mole), natural abundance (%), half-life (s), 100-group neutron capture/activation cross-sections (b), activation product, daughter product and gamma factor (mSv m^2/h MBq) was created.[11,12] The 100-group neutron activation cross-sections were generated using the GROUPR module of NJOY2016 and ENDF neutron cross-section libraries.

For each naturally occurring nuclide within a sample material, the Radioactivity Calculator will determine up to 25 additional nuclides that result from three stages of activation and associated decays. (It should be noted that only the daughter product with the highest branching ratio was considered, and the calculator does not include meta-stable phases or spontaneous fission events.) This activation-decay chain is represented in Figure 8. While decay and activation processes can be infinite, the purpose of this calculator is to obtain a reasonable estimate of the sample radioactivity after a finite amount of time; therefore, only three activation and decay stages were incorporated.

To calculate the radioactivity of a nuclide, the 100-energy group neutron flux ϕ values (from the NDC) and irradiation time t (user input or output from the NDC) are imported. The calculator then solves the number density differential equation associated with each nuclide in the activation-decay chain using the associated 100-energy group neutron capture cross-sections σ (Equations 11 and 12). The radioactivity A is then determined by multiplying the number density N by the associated decay constant λ of the nuclide (Equation 13).

$$\frac{dN_2}{dt} = N_1\sigma_1\phi - \lambda_2 N_2 - N_2\sigma_2\phi \tag{11}$$

$$N_2 = \frac{N_1\sigma_1\phi}{\lambda_2 + \sigma_2\phi}\left[1 - e^{-(\lambda_2+\sigma_2\phi)t}\right] \tag{12}$$

$$A_2 = \lambda_2 N_2 \tag{13}$$

The calculator is also able to determine the gamma dose rate in units of mrem/hr at 30 cm from the source using the following equation and conversion factor (Equations 14 and 15).[13] Based on these equations, the dose rate is only a representation of the gamma radiation being emitted by the sample material. Thus, the calculated dose rate is an underestimate of the true dose rate.

$$\dot{E} = \frac{A\Gamma}{r^2} \tag{14}$$

$\dot{E}$ = Effective dose rate (mSv/hr)

A = Activity (MBq)

Γ = Gamma Factor $\left(\frac{\text{mSv m}^2}{\text{hr MBq}}\right)$

r = Distance from source (m^2)

$$\frac{(\text{Bq})\left(\frac{\text{MBq}}{1x10^6\text{Bq}}\right)\left(\frac{\text{mSv m}^2}{\text{hr MBq}}\right)\left(\frac{1\text{ Sv}}{1000\text{ mSv}}\right)\left(\frac{100\text{ rem}}{1\text{ Sv}}\right)\left(\frac{1000\text{ mrem}}{1\text{ rem}}\right)}{(0.3\text{ m})^2} \quad (15)$$

$$= \frac{\text{mrem}}{\text{hr}} \text{ 30 cm from the source}$$

The radioactivity calculator was originally designed in MATLAB (R2012b) and will be converted to C# for deployment in the CoMET system.

III.C. Results

As discussed above, the calculator requires some user provided data including the sample mass (g) and composition (elements and mass percent). The neutron flux (neutron/cm^2 s) and irradiation time (days) are provided by the NDC based on the reactor and position selected. The calculator returns the radioactivity after a sample has been removed from the reactor for 0 seconds, 1 hour, 1 month, 3 months, 6 months, and 1 year in both Bq and Ci and the gamma dose rate at 30 cm after those same time periods in mrem/hr.

III.D. Validation

Several online radioactivity calculators do exist; however, each one only considers the radioactivity from the first activation stage.[14–16] Due to the high neutron flux and long irradiation times of most experiments inserted into ATR, these calculators are insufficient. While INL experimental analysts use the ORIGEN code to estimate activities, the Radioactivity Calculator described here is intended to be an easily accessible tool for NSUF users during the conceptual design phase of their experiments. In order to improve radioactivity estimates over existing calculators, yet stay accessible to non-experts, three activation stages were considered. Since none of the other online calculator considers three stages, it was difficult to validate the results. However, the mathematics behind the calculator returned comparable results to the other calculators for the first activation stage, and function-shape plotting confirmed the results for subsequent stages. Future validation with actual sample materials and ORIGEN are under consideration.

III.E. Impact

The intent of the Radioactivity Calculator was to improve radioactivity estimates over existing calculators, while creating an easily accessible interface for researchers to use while writing their proposals. Radioactivity estimates generated by the calculator will be used by NSUF technical staff to ensure that PIEs are scheduled at the appropriate facility and will inform workers of any potential safety concerns.

An example of the calculator's significance can be seen with stainless steel. Five activation-decay chains are provided below, each begin with a naturally occurring nuclide found within stainless steel. If only the first activation stage

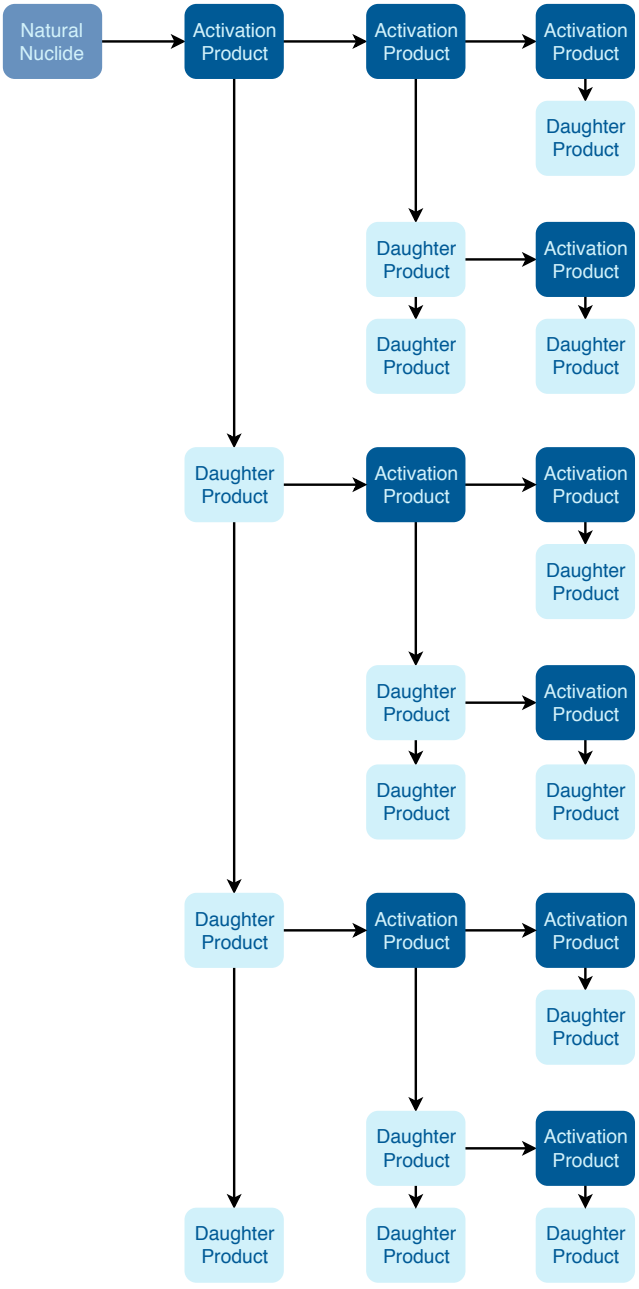

Fig. 8. Activation-decay chain for naturally occurring nuclides utilized in the Radioactivity Calculator

is considered, one of the five activation-decay chains results in cobalt-60. Whereas, if multiple activations (σ) and decays (λ) are considered, all five result in cobalt-60. Cobalt-60 is highly radioactive and has a half-life of over five years. Being able to estimate the radioactivity of stainless steel, which has the potential to activate to cobalt-60, is beneficial to worker safety, facility selection and project schedules.

$$^{59}Co \overset{\sigma}{\Rightarrow} {}^{60}Co$$

$$^{58}Ni \overset{\sigma}{\Rightarrow} {}^{59}Ni \overset{\lambda}{\Rightarrow} {}^{59}Co \overset{\sigma}{\Rightarrow} {}^{60}Co$$

$$^{58}Fe \overset{\sigma}{\Rightarrow} {}^{59}Fe \overset{\lambda}{\Rightarrow} {}^{59}Co \overset{\sigma}{\Rightarrow} {}^{60}Co$$

$$^{58}Fe \overset{\sigma}{\Rightarrow} {}^{59}Fe \overset{\sigma}{\Rightarrow} {}^{60}Fe \overset{\lambda}{\Rightarrow} {}^{60}Co$$

$$^{57}Fe \overset{\sigma}{\Rightarrow} {}^{58}Fe \overset{\sigma}{\Rightarrow} {}^{59}Fe \overset{\sigma}{\Rightarrow} {}^{60}Fe \overset{\lambda}{\Rightarrow} {}^{60}Co$$

IV. DISCUSSION AND CONCLUSIONS

Presented in this paper are two components, the Neutron Damage Calculator and the Radioactivity Calculator, of the CoMET online expert system. These tools will be utilized by NSUF users as they work through the proposal writing and experimental design processes. The goal of these calculators is to enhance researcher proposals and inform technical staff of desired experimental conditions and potential safety and scheduling issues. Both tools use a variety of available nuclear codes and data libraries in order to perform the behind-the-scenes calculations. The data flow associated with these two calculators is presented in Figure 9.

While this project may not be novel in method or approach, the implementation of these two calculators into the broader expert system is a new and highly sought-after concept. Each of these calculators also simplifies the information required from users in order to obtain the needed proposal information, which will assist in the planning and execution of NSUF projects.

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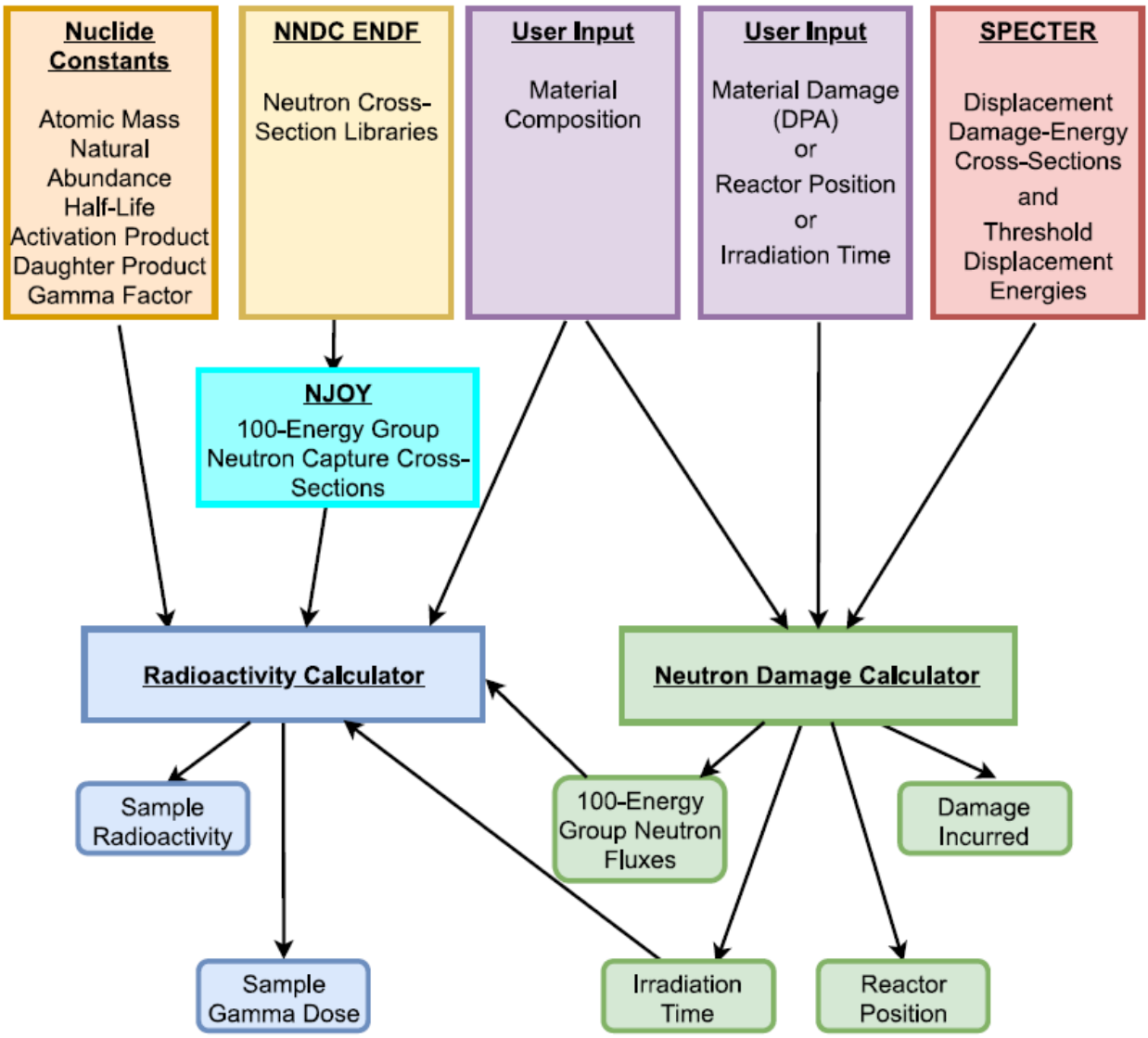

Fig. 9. Data flow diagram for the Radioactivity Calculator and the Neutron Damage Calculator