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Extending the energy range of materials activation modelling

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Abstract

Activation calculations are an essential contribution to understanding the interactions of fusion materials with neutrons. The existing state-of-the-art tools such as EASY-2003 enable calculations to be carried out with neutrons up to 20 MeV. Plans to expose fusion components to high neutron fluxes include the IFMIF materials testing facility. This accelerator-based device will produce neutrons with a high-energy tail up to about 55 MeV. In order to carry out activation calculations on materials exposed to such neutrons it is necessary to extend the energy range of the data libraries. An extension of the European Activation System (EASY) to a new version, EASY-2004, for testing has been completed. The existing reactions have been extended up to 60 MeV and new classes of reactions added using calculated cross sections. Results of preliminary calculations in an IFMIF relevant neutron field are given.

1. Introduction

Understanding and predicting the effects of neutron irradiation on materials is a fundamental requirement of a fusion technology programme. Fusion devices generate fluxes of high-energy neutrons; the activation produced by them influences the planning of maintenance, studies of safety and consideration of waste disposal. Within the EU the response has been the development of the European Activation System (EASY) [1]. In common with previous ones, the current version (EASY-2003) is restricted to neutron energies up to 20 MeV. This energy range is sufficient for fusion devices, but there are plans for facilities to carry out neutron irradiations to test the materials that will be used in future power plants. The most important of these is for the IFMIF device [2], which is an accelerator-based d-Li neutron source able to provide a neutron flux equivalent to 2 $MW\,m^{-2}$, in a volume of 500 cm^3. The neutron spectrum is close to that found in a D–T fusion device, except that there is a high-energy tail of neutrons extending up to about 55 MeV. To include such high-energy neutrons in activation calculations it is necessary to develop EASY further: extending the energy range of the cross section libraries, including data for additional radionuclides and enhancing the FISPACT inventory code. This is a major task and it is being carried out over a two-year period; this paper reports progress at the halfway point with the production of EASY-2004. This is a version for testing, which demonstrates the ability to produce the much larger data libraries and to allow calculations of activation to be made in extended energy neutron spectra.

This paper will describe firstly the capabilities of EASY-2003, secondly the extensive work carried out to validate EASY-2003 is discussed, and then the development of EASY-2004 is covered. Finally some preliminary calculations in an IFMIF spectrum are described.

2. EASY-2003

EASY-2003 consists of the EAF-2003 nuclear data libraries and the FISPACT-2003 inventory code. Part of the documentation [1] gives an overview of the

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system. All EASY-2003 documentation can be downloaded from the web at http://www.fusion.org.uk/EASY2003. EAF-2003 contains cross section data of neutron-induced reactions for energies between 10^{-5} eV and 20 MeV. The pointwise library has increased only slightly in size compared to the previous version (128 MB compared to 126 MB), but has gained considerably in quality and completeness. EAF-2003 contains data for 98 elements from H to Fm, but excludes data for the short-lived radioactive elements At and Fr. There are 774 target isotopes, including ground, first and second isomers, which have non-zero cross sections (i.e. greater than 10^{-8} b) below an energy of 20 MeV. Cross sections to and from isomeric states are listed separately. This leads to a total of 12 617 reaction channels that contain data. Note that a file of uncertainty information for all reactions is included, enabling uncertainty estimates to be calculated by FISPACT. A summary of the EAF libraries is given in [1]. There are reports on cross sections [3], decay data [4] and biological hazard data [5].

FISPACT-2003 [6] is an inventory code that has been developed for neutron-induced activation calculations for materials in fusion devices. However, it can be used with any type of neutron spectrum and is not restricted to only fusion applications. The current version gives extended output, has updated physical constants and corrects minor bugs. It is available for both Windows and UNIX operating systems. Under Windows there is an additional application – the EASY-2003 User Interface – that enables all nuclear data to be viewed and plotted interactively.

The production of the EAF data libraries is a complex task. Several automated systems to aid this have been produced, culminating in SAFEPAQ-II [7,8]. When first introduced this had a radical design that stored all data in relational databases, ran under Windows and was built using Visual Basic. The use of relational databases using the industry standard SQL query language ensured separation between the data and code for the user interface. The use of Windows means that the application is portable. Visual Basic is a fast and convenient way of building the application and is able to use the SQL queries very easily.

Cross section data from almost 60 sources are read and converted from standard formats to databases containing about 4.0 GB of information. By a series of manipulations these data are selected, modified and processed to yield SAFEPAQ-II databases containing about 1.5 GB. All reactions can be graphically displayed, overlaid with experimental data and uncertainty estimates. The experimental data are taken from the EXFOR repository (on CD-ROM) and a custom database. A full log of all changes made to the data is kept, ensuring that a full audit is available.

3. Validation of EASY-2003

In order for trust to be placed on the results of activation calculations it is necessary that the inventory code and the data libraries are validated. By this it is meant that the predictions of the code system are compared with activation measurements made on materials relevant to fusion technology in well-characterised neutron fields. In an ideal world there would be perfect agreement between the two (Experiment (E) and Calculation (C)), but because of uncertainties in the measurements and errors in the data libraries typically a range of C/E values is found.

By measuring a large range of materials it is possible to cover a wide range of reactions, although it must be noted that with the current facilities, it is only possible to validate reactions on stable or long-lived targets that give products with short to medium half-lives. By considering the C/E values it is possible to indicate which reactions are 'validated' – judged to be well described in the library and therefore to be relied on when making predictions of activation. Other reactions with C/E values far from 1 need to be improved before they can be relied on for prediction. Such cross section modifications can be made with confidence if integral results in several complementary neutron spectra are available and if adequate differential data exist. It is then possible to either renormalise the cross section over the entire energy range or renormalise over particular energy regions to get a better fit to the measurements.

Results from the integral measurements have been used with various versions of EAF to validate data relevant for particular materials; also they have provided feedback to aid in the production of EAF-99 to EAF-2003. Summary reports detailing the validation of EASY-2001 and -2003 has been produced [9,10]. To-

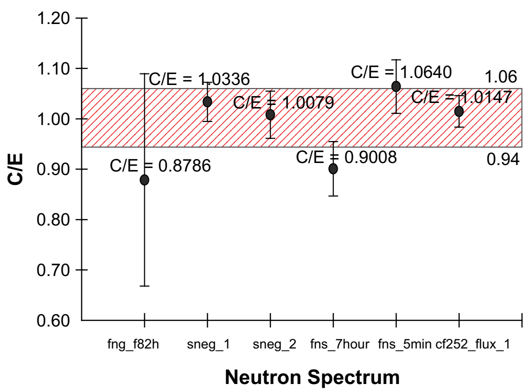

Fig. 1. Ratio of EAF-2003 to measured effective cross sections (C/E) for a series of six integral measurements made in various neutron spectra for the $^{51}V(n,\alpha)\,^{48}Sc$ reaction. The shaded band represents the uncertainty of the cross section in the library.

gether they present all the integral measurements made as part of the European Technology programme, the analysis of Japanese measurements, and list all available experimental data for historical measurements in the ^{252}Cf spontaneous fission spectrum. In total 287 reactions are considered, of these 171 are considered validated. An example of the C/E results for a validated reaction is shown in Fig. 1, the shaded band represents the library uncertainty (typically based on the spread of available differential measurements) and the error bars show the experimental uncertainty for each integral measurement.

4. Development of EASY-2004

There are several issues that must be addressed in order to perform activation calculations at high energies:

- there needs to be a source of cross section data at the higher energies;
- SAFEPAQ-II needs extension to be able to either handle conventional data or data up to 60 MeV;
- the number of nuclides in the decay data library must be increased;
- new group structures must be defined;
- the existing reactions must be extended to 60 MeV;
- new classes of reactions (with thresholds above 20 MeV) must be defined;
- FISPACT must be extended to use the new data.

Although there are some measurements and evaluated libraries with data >20 MeV, these are rather sparse and for a source of cross section data at high energies it is necessary to rely on calculation. There is a major code development project underway led by A Koning involving the TALYS code system [11]. This is a very versatile system enabling the production of a wide range of data using a unified set of models. As part of this development a series of libraries was produced containing activation cross sections for energies up to 60 MeV. Feedback on early libraries enabled many bugs and problems with the code to be removed. The data library termed TALYS-4 was read into SAFEPAQ-II and was used as the basis for the production of EAF-2004.

The decision was made to retain the same method of reaction description as used in earlier EAF libraries – use of MT numbers. Because of the new reaction classes that are required some unofficial extension of the ENDF MT numbering system was required and some 49 new MT values were defined (occupying unused values between 152 and 200) and used by both TALYS and SAFEPAQ-II. This approach is feasible for energies up to 60 MeV but would probably be too complicated for higher energies. The approach taken with the IEAF-2001 library [12] of using the total cross section and values for the production of a range of product nuclides would then be necessary.

SAFEPAQ-II has required significant changes to allow the new data to be viewed and to enable the TALYS-4 data to be automatically 'joined' at 20 MeV to the existing data. Many other new features have also been introduced to make the handling of the high-energy data easier.

The additional reactions mean that new nuclides can be formed, and these require entries in the nuclear data library. The number of nuclides has increased from 1917 in EAF-2003 to 2195 in EAF-2004.

EAF-2004 consists of a point-wise library extending to 60 MeV, however this requires processing into multi-group form before it can be used in application such as FISPACT. Existing group structures extend up to 20 MeV, so some extension was required for the new data. The existing 175 and 315 group structures have been extended by adding groups of width 1 MeV up to 55 MeV.

The basic method of production of EAF-2004 has involved the addition of new reactions (generally with thresholds >20 MeV) taken directly from TALYS-4. For the reactions already present in EAF-2003, the TALYS-4 data above 20 MeV were scaled to smoothly fit the existing data <20 MeV. In some cases very large factors were required and for many of these it is unphysical to use the very limited data in EAF-2003 as a basis and it is preferable to take all the data from TALYS-4.

FISPACT has been modified so that data in the new group structures can be read. The pathway facilities have been extended to include the new reaction classes and the many arrays have been enlarged to accommodate the much larger amount of data. In previous EAF libraries, uncertainty data was available in one group for threshold reactions and in three groups for (n,γ) and (n,f) reactions. A new group has been defined for 20–60 MeV, meaning that threshold uncertainty data are now in two groups and all non-threshold reaction (not just the two previously mentioned) uncertainties are in four groups. Changes in FISPACT allow this new uncertainty structure to be read and used.

The statistics for EAF-2004 are: 62 860 reactions on 775 targets from ^{1}H to ^{257}Fm in the energy range 10^{-5} eV to 60 MeV, the pointwise library has a size of 184 MB.

An alternative source of nuclear data for activation above 20 MeV is the IEAF-2001 library [12] developed by FZK. This has been used with the ALARA [13] and ANITA [14] code systems, but these are not discussed here because of space limitations.

5. Preliminary activation calculations

EASY-2004 is designed as a 'proof-of-principal' version; it is not designed for distribution to users. It will be used for extensive testing and improvement prior to the production of EASY-2005. However, EASY-2004 is able to carry out preliminary calculations that demonstrate the ability to make predictions on irradiation of materials in IFMIF.

As an example, consider the irradiation of Eurofer steel for five years in the high flux test region (HFTM) of IFMIF. A calculation was made with the complete neutron spectrum (211 groups) and using a flux of 7.5×10^{14} $ncm^{-2}\,s^{-1}$. A second calculation removed the neutrons above 20 MeV (6.5%) and so used 175 groups and a flux of 7.0132×10^{14} $ncm^{-2}\,s^{-1}$. The results of the activity from these two calculations are shown in Fig. 2. The nuclides which dominate the activity at various times (symbol is at the nuclide half-life with a value equal to the activity at shutdown) are plotted. At times up to 10 years there is a difference of about 40% between the two cases, dropping to about 18% at 10 000 years. This is not due to different nuclides being formed but rather to the fact that more of each nuclide has been formed in the 211-group case. This is either due to the high-energy neutrons increasing the contribution of existing reactions or to new reactions becoming important at the higher energies.

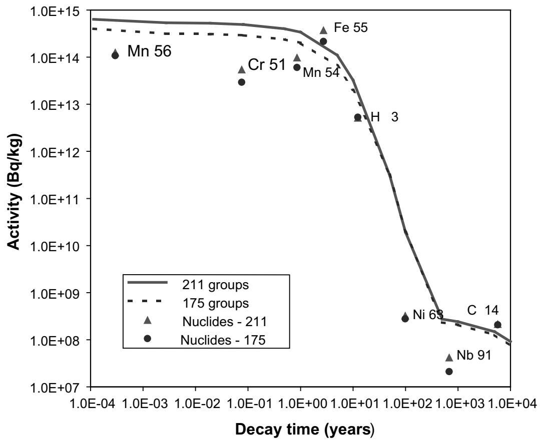

Fig. 2. Activity as a function of time after shutdown in Eurofer irradiated in an IFMIF spectrum. '211 groups' refers to the full neutron spectrum, while '175 groups' has the neutrons from 20–55 MeV removed. The symbols show the dominant nuclides, plotted at the position of half-life and activity at zero time.

Some examples of the contributions of reactions in the two cases, including some of the new reactions, are shown in Table 1. Note that in Table 1 reaction labels such as (n,pα) are shorthand for (n,pα + nph + dh). The high-energy neutrons are responsible for increasing the number of nuclides listed at shutdown from 580 to 823. However, it is interesting that such an irradiation results in very few new dominant nuclides (some like ^{48}V, ^{49}Sc contribute at the sub 1% level) and to only a minor contribution from the new reactions. This suggests that future effort on improving the library should be on the existing reactions rather than on the new reaction classes.

6. Conclusions

The current tool for activation calculations, EASY-2003 is described. This is a comprehensive set of nuclear data libraries (EAF) with the inventory code FISPACT. It has been validated by a series of measurements made in fusion relevant neutron spectra. The current version of EASY contains data up to 20 MeV, which is sufficient for calculations on fusion devices but not for materials test facilities such as IFMIF. Work to extend EASY to 60 MeV is reported and has resulted in the production of a test version, EASY-2004. This contains a large number of reactions (62 860 compared to 12 617) and additional nuclides (2200 compared to 1917). This version is for internal testing prior to the next release version, EASY-2005, that is expected to be available at the beginning of 2005.

Table 1
Examples of reactions in EAF-2004 and their contributions to the activity of Eurofer following irradiation in the high flux region of IFMIF, due to various nuclides in the 175 (10^{-5}–2×10^{7} eV) and 211 (10^{-5}–5.5×10^{7} eV) group cases

Reaction	Threshold (MeV)	Activity at shutdown (Bq) – 175 groups	Activity at shutdown (Bq) – 211 groups
^{54}Fe(n,p)^{54}Mn	–	5.92×10^{13} (97.5%)	6.11×10^{13} (62.7%)
^{56}Fe(n,p)^{56}Mn	2.97	1.07×10^{14} (99.0%)	1.23×10^{14} (96.0%)
^{56}Fe(n,pα)^{52}V	11.00–31.95	0	5.82×10^{11} (5.0%)
^{56}Fe(n,2n)^{55}Fe	11.40	2.15×10^{14} (99.4%)	3.64×10^{14} (98.0%)
^{56}Fe(n,t)^{54}Mn	12.66–20.78	0	3.42×10^{13} (35.1%)
^{54}Fe(n,dα)^{49}V	16.08–37.04	0	1.69×10^{11} (7.4%)
^{57}Fe(n,3n)^{55}Fe	19.18	0	5.87×10^{12} (1.6%)
^{56}Fe(n,2nα)^{51}Cr	20.00–48.81	0	3.17×10^{12} (5.8%)
^{50}Co(n,3α)^{39}Ar	25.89	0	2.21×10^{4} (13.4%)
^{183}W(n,5n)^{179}W(β^{+})^{179}Ta	29.51	0	1.03×10^{10} (3.1%)

Preliminary calculations of the activation of Eurofer in an IFMIF spectrum (containing 6.5% of neutrons with energy >20 MeV) using EASY-2004 show that the additional neutrons result in increased activation (40–15%) over times up to 10 000 years after shutdown. The nuclides dominating the response are the same as for the calculation with no high-energy neutrons and the reactions are also similar, although some with high thresholds not present in EASY-2003 make significant contributions. Future work will involve the validation of EASY-2004 with the available experimental data (including some with neutrons >20 MeV) and feedback from the validation of EASY-2003 to improve EAF. In addition, extra features, such as the ability to read the existing IEAF-2001 library, will be added to FISPACT. The improvements that will be made during production of EASY-2005 mean that it should give improved calculations with neutrons both above and below 20 MeV.

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