# Covariances for the $^{56}$Fe radiation damage cross sections

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**Abstract.** The energy-energy and reaction-reaction covariance matrices were calculated for the n +$^{56}$Fe damage cross-sections by Total Monte Carlo method using the TENDL-2013 random files. They were represented in the ENDF-6 format and added to the unperturbed evaluation file. The uncertainties for the spectrum averaged radiation quantities in the representative fission, fusion and spallation facilities were first time assessed as 5–25%. Additional 5 to 20% have to be added to the atom displacement rate uncertainties to account for accuracy of the primary defects simulation in materials. The reaction-reaction correlation were shown to be 1% or less.

## 1. Introduction

The goal of this work is an evaluation of the covariance matrices for the physical quantities used for characterization of the neutron induced radiation damage in the materials. They include: the kinetic energy released by charged particles KERMA (locally deposited nuclear heating), damage energy (eventually defines the number of displaced atoms) and gas production cross sections (n,x$\alpha$), (n,xt), (n,xp) …(predict transmuting of target nuclei into gases).

The uncertainties and energy-energy or reaction-reaction correlations for these quantities were not assessed so far, whereas the covariances for many underlying cross sections are often presented in the evaluated data libraries.

Since damage quantities depend on many reactions channels and energy-angular distributions of reaction recoils, the evaluation of uncertainty is not straightforward. To reach a declared goal, we used an idea of the Total Monte Carlo application to Nuclear Data [1].

This paper summarises the results for evaluation, validation against measurements and representation in the ENDF-6 format of the n +$^{56}$Fe radiation damage covariances from thermal energy up to 20 MeV. This study was motivated by the IAEA Coordinated Research Project "Primary Radiation Damage Cross Sections" [2]. Preliminary results were reported at the IAEA Technical Meeting "Nuclear Reaction Data and Uncertainties for Radiation Damage" held in June 2016 [3].

## 2. Method of evaluation of energy-energy and reaction-reaction covariances

We used one unperturbed (original) and five hundreds randomly perturbed evaluated files for n +$^{56}$Fe reaction from TENDL-2013 evaluations [4]. The random files were generated by sampling of the input underlying model parameters within their uncertainties, which represent the spread of known experimental cross section and nuclear structure data.

All 501 files were processed by the NJOY-2012.50+ code [5] to calculate the damage quantities of interest in the ENDF-6 format. The modules RECONR and BROADR were used to reconstruct the cross sections at room temperature 293$^o$K, HEATR and GASPR - to calculate KERMA (designated by MT = 301), Damage Energy (MT = 444) and gas production cross sections (n,x$^4$He) (MT = 207), (n,x$^3$He) (MT = 206), (n,xt) (MT = 205), (n,xd) (MT = 204) and (n,xp) (MT = 203). Finally GROUPR was used to generate desired data in the grouped-wise format *gendf* to reduce the number of points but still representing the characteristic structures in the cross sections. For this the VITAMIN-J 175-groups representation was employed, which covers the energies from $10^{-5}$ eV to 19.64 MeV.

A Fortran-90 code was written to read the NJOY output *gendf* files and calculate the mean (averaged over 500 random evaluations) quantities, energy-energy (E-E) and reaction-reaction (MT-MT) correlations matrices.

Following the general definitions [6], the first order covariance matrix for values of function $y_i$ was calculated from the $N_{random}$ random set as

$$cov\left(y_i, y_j\right) = \sum_{N_{random}} \frac{\left(y_i - \bar{y}_i\right)\left(y_j - \bar{y}_j\right)}{N_{random}}, \quad (1)$$

where indices $i$ or $j$ refer to the quantities for the specific energy group or reaction MT, $\bar{y}_i$ is an averaged value.

The diagonal elements $(i = j)$ of covariance matrix deliver a variance or square of the standard deviation $\sigma_i$:

$$\sigma_i^2 = cov\left(y_i, y_i\right) \quad (2)$$

The correlation matrix was then calculated as:

$$cor\left(y_i, y_j\right) = \frac{cov\left(y_i, y_j\right)}{\sigma_i \sigma_j}, cor\left(y_i, y_i\right) = 1 \quad (3)$$
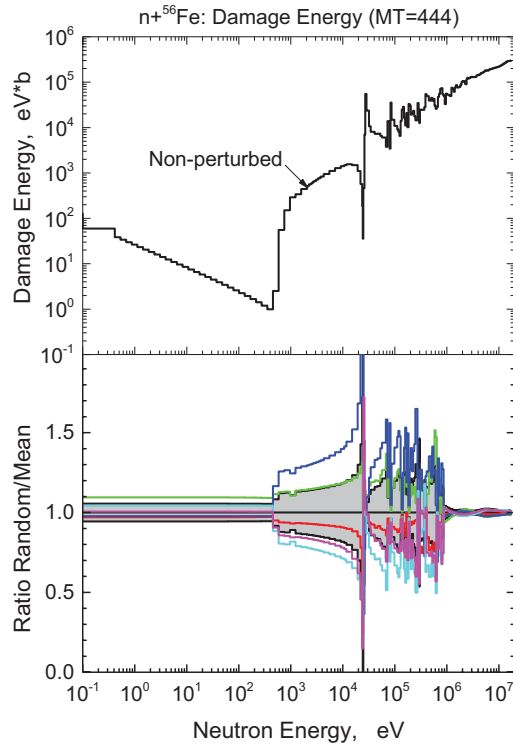
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**Figure 1.** Damage energy: (top) non-perturbed TENDL-2013 evaluation; (bottom) ratio of 5 random files (colour curves) to mean and standard deviation (grey zone).
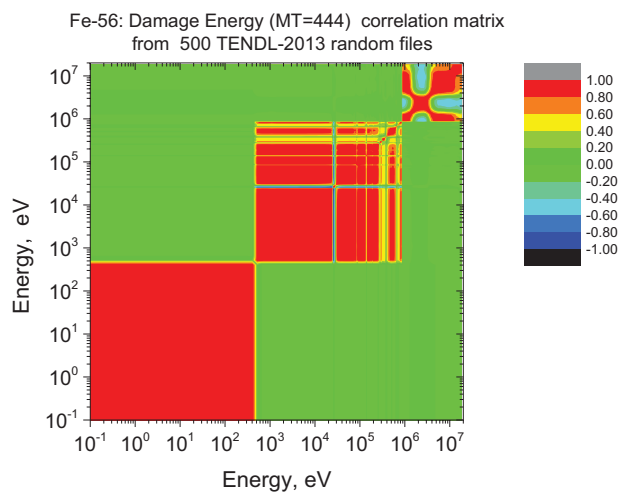


**Figure 2.** Energy-energy correlation matrix calculated from 500 TENDL-2013 random files.

## 3. Derived covariance matrices

As an example of calculations, Figs. 1 and 2 display the damage energy, its uncertainties and energy-energy correlation matrix for neutron interaction with $^{56}$Fe.

Analysing obtained covariance data we observed:

– Damage energy (MT = 444) – 1 to 40% uncertainties and strong positive energy-energy correlations (coefficients $\leq 1$) inside 2–3 large domains which do not correlate each other;
– KERMA (MT = 301) – similar to damage energy but uncertainties range from 2 to 20%;
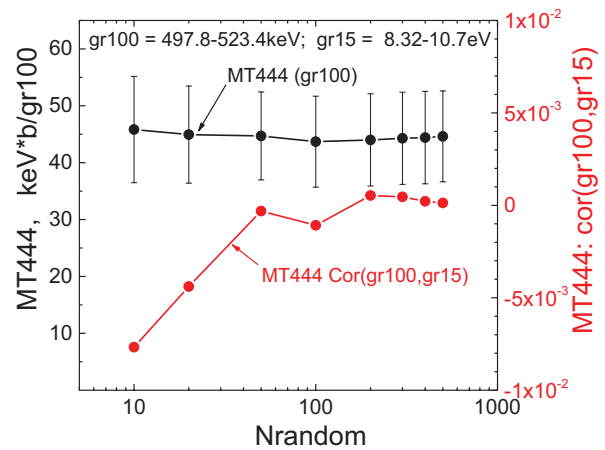


**Figure 3.** Damage energy and uncertainty for energy group #100, and its correlation with #15 versus $N_{random}$.
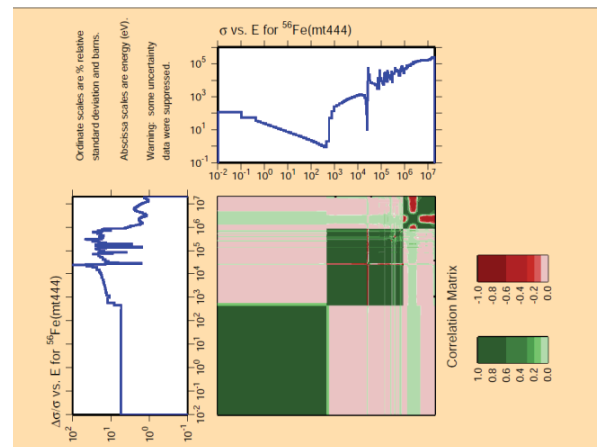


**Figure 4.** Damage energy, its relative uncertainty and correlation matrix for n-$^{56}$Fe processed and plotted by NJOY-2012.

– He-4 production (MT = 207) – $\approx 30\%$ uncertainties and positive energy-energy correlations in the whole energy range where the correlation strength gradually decreases from 1 to 0.

The statistical significance of the covariance matrix elements was checked. As an example, Fig. 3 shows the mean value of damage energy (MT444) for energy group #100 (497.8–523.4 keV), its uncertainty and correlation coefficient between this group and #15 (8.32–10.7 eV) versus the number of the random files $N_{random}$ used for calculation of these values. It is seen that the mean value and its uncertainty stabilize for ensemble of 50–100 sampled files, whereas the correlation coefficient converges when 200–300 files are used.

The derived covariance matrices for MT203-444 were converted in the ENDF-6 formatted file MF33 and were checked for positive definiteness computing the eigenvalues by code COVEIG [7]. Then MF33/MT203-444 data were added to complete TENDL-2013 evaluation for $^{56}$Fe. The compliance with ENDF-6 format rules was proved by processing with standard checkers and covariance relevant modules ERRORR and COVR of NJOY-2012. Fig. 4 shows the NJOY plots for the damage energy covariance matrix MF33/MT444.
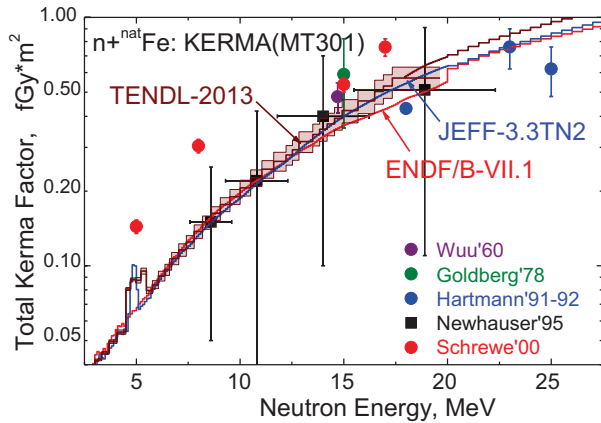
**Figure 5.** The KERMA factor for Iron: comparison of TENDL-2013 (including uncertainties) with other evaluations and measured data.
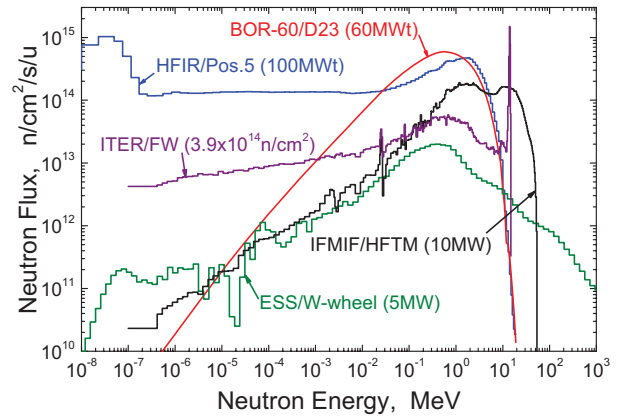


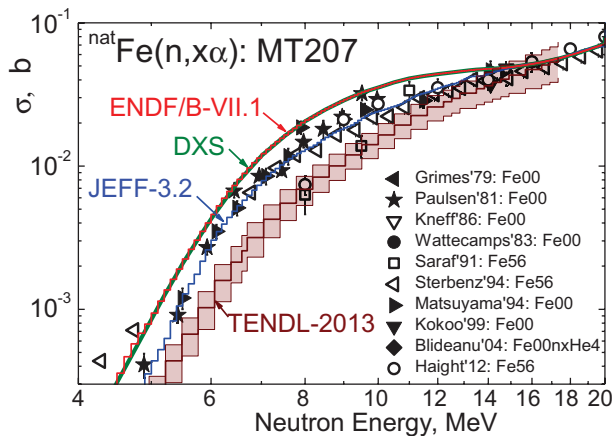**Figure 7.** Neutron energy spectra in the fission, fusion and spallation nuclear facilities.



**Figure 6.** $^4$He production cross section for Iron: comparison of TENDL-2013 (including uncertainties) with other evaluations and measured data.

## 4. Comparison of the evaluated uncertainties with experiment

The evaluated uncertainties were compared with known measured data. It is possible to do for the KERMA factors and (n,x$\alpha$) cross sections, Figs. 5 and 6. It is seen that calculated energy dependant standard deviation, which varies from 5 to 20%, are comparable with uncertainties or spread of measured data and differences between major evaluations.

## 5. Energy averaged damage quantities

To demonstrate the practical importance of obtained covariance data we computed the energy averaged damage quantities for $^{56}$Fe inside the representative fission, fusion and spallation nuclear facilities. Their energy spectra are displayed in Fig. 7.

For the actual calculations we selected: (1) thermalized spectrum in centre of the irradiation channel C5 inside the High Flux Isotope Reactor HFIR (HFIR/C5); (2) fast spectrum with 14-MeV peak in the First Wall of ITER (ITER/FW); (3) spectrum with high cut-off energy 55 MeV averaged over the High Flux Test Module volume of the

projected accelerator driven Fusion Material Test Facility IFMIF (IFMIF/HFTM).

The neutron fluxes during a full operation in these facilities and calculated damage quantities for $^{56}$Fe including uncertainties are summarised in Table 1. The uncertainties were computed with both full energy-energy covariance matrices and also, for comparison, only with diagonal elements. As seen the ignorance of the off-diagonal correlations results to the underestimation of uncertainties by a factor 2–3.

Table 1 also lists the reaction-reaction (MT-MT) correlations, which are turned to be very small. Thus for most practically important correlation between NRT-dpa and He-production (this ratio is used as scaling parameter for the fusion materials), the correlation coefficient is less than $2 \times 10^{-4}$. The correlations between different gases production rates, such as (n,xp), (n,xa), (n,xt), are below $10^{-2}$.

The contributions from the Nuclear Data and Material Physics to the total uncertainties for nuclear heating, dpa and gas production in the representative nuclear facilities are compared in Table 2. The Nuclear Data are the sole source for the damage energy and gas production. On the other hand, the Material Physics contributes to the NRT-dpa as the uncertainty of the lattice threshold $E_d$. For the iron crystalline lattice, an averaged $E_d$ was estimated to be $40 \pm 2$ eV or $\pm 5\%$ [8].

The athermal recombination-corrected *arc*-dpa additionally depends on a simulation of the primary defects surviving function in frame of Molecular Dynamics or Binary Collision Approximation. The "OECD fit" to the surviving efficiency estimates its uncertainty as $\pm 2\%$ [9]. However the visible spread of the MD results is essentially larger and should be increased up to about $\pm 20\%$.

## 6. Conclusion

The Total Monte Carlo method and 500 TENDL-2013 random evaluated files for the n+$^{56}$Fe reaction were used to qualify the energy-energy and reaction-reaction covariance matrices of the radiation damage cross sections up to 20 MeV, i.e. the nuclear heating due to the charged particles, damage energy, NRT- or *arc* -atom displacements and gas production cross sections.

**Table 1.** The spectrum averaged damage quantities and their uncertainties assessed for $^{56}$Fe under neutron irradiation in HFIR/C5, ITER/FW and IFMIF/HFTM (<20 MeV). *Italic font* indicates the calculated uncertainties when off-diagonal elements of covariance matrices are ignored.

| Facility/Location | HFIR/C5 | ITER/FW | IFMIF/HFTM |
|---|---|---|---|
| Neutron Flux[n/cm$^2$/s] | 5.10E+15 | 3.90E+14 | 7.32E+14 |
| Averaged Energy [MeV] | 0.41 | 3.47 | 7.00 |
| Displacements [dpa/fpy] | 30.5 ± 0.95 (3.1%) | 10.4 ± 0.16 (1.5%) | 25.1 ± 0.36 (1.4%) |
| | *30.5 ± 0.24 (0.8%)* | *10.4 ± 0.05 (0.5%)* | *25.1 ± 0.09 (0.4%)* |
| KERMA [W/Kg] | 277 ± 7 (2.7%) | 246 ± 21 (8.7%) | 469 ± 31 (6.7%) |
| | *277 ± 2 (0.7%)* | *246 ± 12 (4.9%)* | *469 ± 10 (2.2%)* |
| (n,x$^4$He) [appm/fpy] | 5.7 ± 1.3 (23%) | 92 ± 21 (23%) | 136 ± 31 (23%) |
| | *5.7 ± 0.3 (6%)* | *92 ± 12 (13%)* | *136 ± 11 (7.8%)* |
| (n,x$^1$H) [appm/fpy] | 37 ± 6.3 (16.9%) | 410 ± 72 (17%) | 642 ± 102 (16%) |
| | *37 ± 1.6 (4.4%)* | *410 ± 41 (10%)* | *642 ± 31 (4.9%)* |
| MT-MT max. correlation | 204–206 = + 1.05% | 205–206 = + 1.07% | 204–206 = + 0.66% |
| between (n,x$^4$He) & (dpa) | 207–444 = −5.9E-5 | 207–444 = + 1.8E-4 | 207–444 = + 1.4E-4 |

**Table 2.** The contribution of Nuclear Data and Material Physics to uncertainties of the spectrum averaged damage quantities assessed for $^{56}$Fe under neutron irradiation in HFIR/C5, ITER/FW and IFMIF/HFTM.

| Source of Uncertainty | Nuclear Data | Material Physics |
|---|---|---|
| Damage Energy DE (MT=444) | ± (1.4 - 3.1)% | do not contribute |
| Lattice Threshold E$_d$ = 40 ± 2 eV [8] | do not contribute | ± 5.0% |
| **Total Uncertainty for    NRT-dpa** = 0.8*DE/2Ed | **± (5.2 - 5.9)%** | |
| Primary Defects Surviving Efficiency | do not contribute | from OECD fit   ≈ ± 2% [9] spread of MD res. ~ ± 20% [9] |
| **Total Uncertainty for   arc-dpa** ~ NRT-dpa*Efficiency | **± (5.2 - 5.9)% + ±20% ≈ 21%** | |
| **KERMA or Nuclear Heating from charged products** | ± (2.7 - 8.7)% | do not contribute |
| **Gas production: (n,xα) or (n,xH)** | ± 23% or ± 17% | do not contribute |

The obtained covariance matrices were merged as MF33 formatted file in the complete unperturbed TENDL-2013 file and were tested for positive definiteness, compliance with ENDF-6 rules and processing by the NJOY code.

For the practical applications, the uncertainties of the energy weighted quantities inside the representative fission, fusion or spallation nuclear facilities were first time calculated. They equal (1.3–3.0)% for NRT-dpa, (3–9)% for KERMA and (17–23)% for gas production. These uncertainties however will decrease by 2–3 times if the diagonal-off elements of the energy-energy correlation matrices are omitted.

The uncertainty for NRT- and *arc*- dpa, estimated from underlying nuclear data, have to be additionally increased by (5–20)% due to additional uncertainties of the involved Material Physics parameters, i.e. the lattice threshold and primary defects surviving efficiency.

Reaction-reaction correlations were shown to be negligibly small: between He and dpa <2 10$^{-4}$, between any gas ($^4$He, $^3$He, t, d, H) production rates <10$^{-2}$.

The dpa and gas production cross sections uncertainties but evaluated differently [10] have close values.

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