



**HAL**  
open science

# Impact of the ageing of materials on primary damage induced by different neutron spectra

David Simeone, L. Luneville

► **To cite this version:**

David Simeone, L. Luneville. Impact of the ageing of materials on primary damage induced by different neutron spectra. 2023. hal-04101542

**HAL Id: hal-04101542**

**<https://hal.science/hal-04101542>**

Preprint submitted on 20 May 2023

**HAL** is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

# Impact of the ageing of materials on primary damage induced by different neutron spectra

L. Luneville<sup>a,1,\*</sup>, D. Simeone<sup>b</sup>

<sup>a</sup>*DEN-Service d'Etudes et de Recherche en Mathématiques Appliquées, LRC CARMEN CEA-CNRS-ECP/SPMS, Université Paris-Saclay, F-91191, Gif-sur-Yvette, France*

<sup>b</sup>*DEN-Service de Recherches Metallurgiques Appliquées, LRC CARMEN CEA-CNRS-ECP/SPMS, Université Paris-Saclay, F-91191, Gif-sur-Yvette, France*

---

## Abstract

The recent nuclear evaluations like the Thalys based Evaluation Nuclear Data Library (TENDL) are now able to accurately estimate either from Monte Carlo simulations or experimental results the many elastic and inelastic neutron-atoms interactions and allow calculating more realistically primary damage induced by nuclear reactions. Even if these calculations do not take into account relaxation processes occurring at the end of the displacement cascade, they can accurately describe primary and recoil spectra in different reactors or ion beam accelerators. Since neutrons are sensitive to isotopes and not to chemical elements, these spectra must be calculated taking into account all neutron-isotopes interactions separately and then be summed over the relative abundance of different isotopes forming chemical elements. Such a procedure is applied in the DART code developed to compare irradiation in different facilities. Such a procedure gives an accurate estimation of primary damage induced by different fluxes within the Binary Collision Approximation using the TENDL-2015 library allowing to mimic primary damage produced in different nuclear facilities with ion beams. In this work we discuss the potentiality of such an approach computing primary damage in Ni based steel springs of nuclear reactors for different irradiation times, pointing out the importance of ageing, i.e. the modification of isotopic abundance of Ni isotopes with the irradiation fluence, on these spectra. This calculation taking into account accurate neutron isotope cross sections is crucial to determine the nature and the energy of ions used in ion beam facilities to mimic the micro structural evolution of actual and future materials for nuclear plants.

*Keywords:* ageing, neutron-isotope cross section, primary and recoil spectra

---

## 1. Introduction

When a material is subjected to a neutron flux, atoms of the solid can be knocked from their equilibrium positions with a wide range of energies. The nature and the energy of recoil atoms produced during inelastic neutron isotopes collisions determine for a large part the primary damage responsible for the long term stability of the material under irradiation. Averaging these recoils energies over the isotopic composition allows studying the impact of ageing, i.e. the variation of the isotopic composition versus the fluence, on the primary damage. The detailed description of calculation performed by the DART code <sup>1</sup> has already been discussed

in detail in previous articles [1] and only specific applications of this code to study the ageing of materials are discussed in this work. To quantify the effect of ageing of materials, different Primary Knocked Atoms (PKA) spectra and displacement cross sections [2, 3] have been computed in nickel-containing alloys with different compositions for different facilities.

The impact of the isotopic composition on the primary damage on Ni element is discussed in detail in the first part of this work pointing out the need to have accurate neutron isotopes cross section describing all possible open channels. This last point implies a large and accurate estimation of all stable and non stable nuclei produced during a neutron isotope interaction. All calculations presented in this work were performed using the TENDL-2015 library which combines Monte Carlo and analyses of experimental measurements to compute these data [8]. A close inspection of all neutron iso-

---

\*I am corresponding author

Email addresses: [laurence.luneville@cea.fr](mailto:laurence.luneville@cea.fr) (L. Luneville), [david.simeone@cea.fr](mailto:david.simeone@cea.fr) (D. Simeone)

<sup>1</sup>available at OECD:

(<http://www.oecd-nea.org/tools/abstract/detail/nea-1885/>)

tope cross sections points out that primary damage, i.e. displacement of atoms induced by the neutron impinging over a time scale smaller than  $10^{-9}$  second, is only produced by  $^{58}\text{Ni}$  and  $^{59}\text{Ni}$  isotopes. The dependence of the PKA spectra and the displacement cross sections as a function of the different isotopes and the neutron fluxes of different facilities is discussed in the second part of this work, allowing then to discuss the effect of aging in Ni based steels.

## 2. Isotopic composition and primary damage

When a neutron interacts with an atom at rest in a crystal, a part of the kinetic energy of the neutron is given to the atom leading to the recoil of the atom (elastic cross section) or a nuclear reaction (inelastic cross section). The cross sections associated with a specific interaction determine the intensity of the process and the part of the energy given to the atom at rest (elastic collision) or to daughter atoms produced during the nuclear reaction (inelastic collision). Obviously, the cross sections associated to each open channel is different for each isotope of a given chemical element. To accurately compute the all possible nuclear interactions produced by impinging neutrons in a given material, all neutron isotopes cross sections must be computed and then averaged according to the isotopic composition of each chemical element forming the crystal. This procedure is applied in the DART code [1] and such a procedure is different from those used in other codes like SPECTER [4] which computes only neutron-element cross sections. Such a procedure thus allows following the evolution of primary damage versus the material ageing, i.e. the variation of its isotopic composition as a function of the dose.

To point out the impact of the isotopic composition on the primary damage for different facilities, we compute primary spectra associated with the energy redistribution of primary knocked on atoms for different facilities and the displacement cross sections versus the energy of the incident neutron, at different isotopic values for pure nickel atoms. The variation of the isotopic composition of  $^{58}\text{Ni}$  versus the irradiation time may give some clues to explain the unexpected embrittlement of Ni based alloys springs observed in Candu reactors [5, 6]

Figure 1 displays the neutron spectra of different nuclear facilities used to study the aging of Ni based alloys. To highlight the effect of inelastic collisions, PKA spectra and displacement cross sections induced by different ion beams (100 keV Xe and 800 keV Ar) were also calculated for pure Nickel.

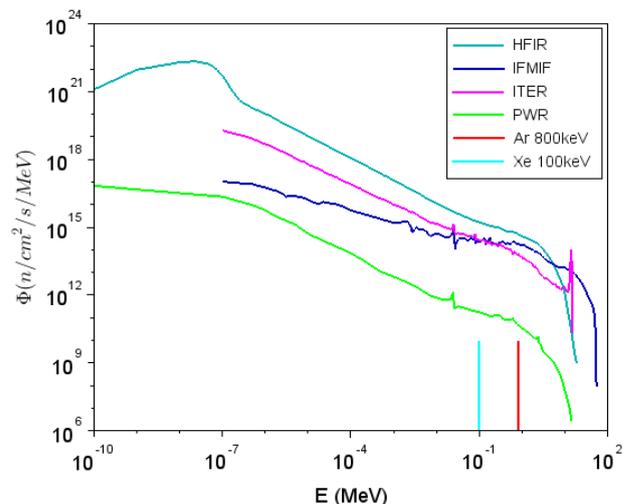


Figure 1: Neutron and ions spectra  $\phi$  for different nuclear reactors and ion beam facilities (pink line: ITER (fusion reactor), turquoise line: HFIR (High Flux Isotope Reactor), navy line: IFMIF (International Fusion Materials Irradiation Facility), green line: PWR (Power Water Reactor); red line: Ar 800 keV; blue line: Xe 100 keV)

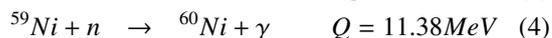
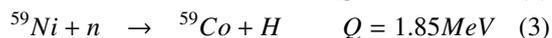
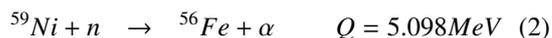
### 2.1. Nuclear cross sections of Ni isotopes

The pure nickel is mainly composed by 3 different isotopes. The  $^{58}\text{Ni}$  is the most abundant isotope (67.8%) followed by  $^{60}\text{Ni}$  (26.2%) and finally the  $^{62}\text{Ni}$  (3.7%). The two last isotopes  $^{60}\text{Ni}$  and  $^{62}\text{Ni}$  are stable and their inelastic neutron isotopes cross section can be neglected. Even if neutron-isotope cross sections are similar for all three isotopes, the  $^{58}\text{Ni}$  is the most abundant isotope and is the only one able to produce the  $^{59}\text{Ni}$  metastable nucleus according to the capture reaction:



The  $^{58}\text{Ni}$  capture cross section follows a square root law of the neutron kinetic energy in the center of mass frame. This capture cross section varies from 100 barns to 0.01 barns for a neutron kinetic energy ranging from  $10^{-5}$  eV to  $10^3$  eV.

The unusual properties of metastable  $^{59}\text{Ni}$  isotope is well established[5, 7]:



The  $^{59}\text{Ni}(n, \alpha)$  reaction releases 5.1 MeV leading to the emission of a 4.8 MeV  $\alpha$  particle and 340 keV  $\text{Fe}$  recoils for thermal neutrons. The second reaction  $^{59}\text{Ni}(n, p)$  mainly generates a 1.85 MeV proton and 31

keV  $^{59}\text{C}$  recoils for thermal neutrons. The last nuclear reaction only induces the emission of a  $\gamma$  which does not participate to the production of primary damage.

## 2.2. Energy spectra of recoil atoms

Neglecting the  $\gamma$  emission, the number of daughter recoils produced during the nuclear reaction depends on the  $n$ - $^{59}\text{Ni}$  capture cross section versus the kinetic energy of the impinging neutrons. Such capture cross section for two distinct channels are given by neutron libraries. In this work, all  $n$ - $^{59}\text{Ni}$  capture cross section were computed from the TENDL-2015 [8] cross section data library which contains the most accurate evaluations on  $^{59}\text{Ni}$ . Obviously, the energy of recoil atoms ( $^{56}\text{Fe}$  and  $^{59}\text{Co}$  in our case) is not only a function of the kinetic energy of the incident neutron but also of the angular distribution of emitted particle in the center of mass frame:

$$T(\mu, E) = \frac{1}{(A+1)}(E^* - 2\mu\sqrt{A'E^*E_a} + A'E_a) \quad (5)$$

where  $E^* = \frac{A+1-A'}{1+A}E$  and  $E_a = Q + \frac{A}{A+1}E$  in the center of mass frame. In these expressions,  $A$  is the mass ratio of incident particles and  $A'$  is the mass ratio of ejected particles [9]. Then it appears that the energy of recoils induced by nuclear reaction listed in Eq. (2) depend on the kinetic energy of incident neutron and then the neutron fluxes of different facilities.

This expression points out that the energy of recoil atoms are function of the kinetic energy of the incident neutron and then PKA spectra induced by different facilities will be function of their neutron flux plotted on Fig. 1. To point out the impact of neutron fluxes on PKA spectra, different facilities associated with fusion and fusion reactors are plotted on this graph. In order to achieve the comparison with ion beam facilities, typical ion beam spectra which behave as Dirac distributions are also plotted on the same graph just for comparison.

## 3. PKA spectra and displacement cross sections

To highlight the impact of different Ni isotopes on the PKA spectra, Figure 2 displays the variation of the PKA spectra as a function of its kinetic energy for  $^{58}\text{Ni}$  and  $^{59}\text{Ni}$  isotopes irradiated in the HFIR reactor [10] used to test materials for fission reactors, exhibiting a large thermal neutron flux (cf Fig. 1). Two distinct peaks clearly appear on the PKA distribution for the  $^{59}\text{Ni}$  iso-

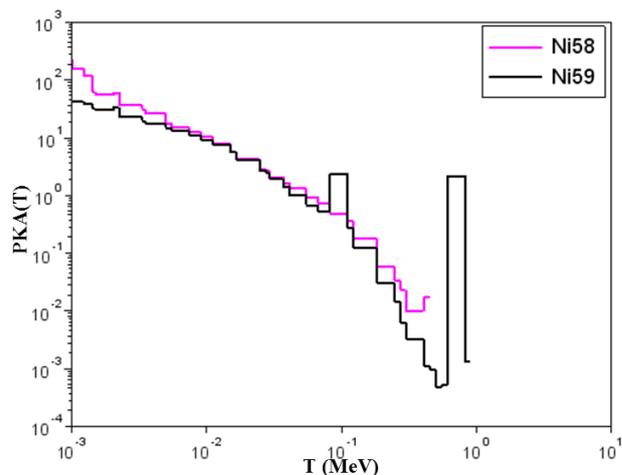


Figure 2: PKA spectra as a function of the energy of recoil atoms  $T$  in pure  $^{58}\text{Ni}$  (magenta line) and  $^{59}\text{Ni}$  (black line) isotopes. Two distinct peaks are clearly observed on the  $^{59}\text{Ni}$  PKA distribution pointing out the impact of the Ni isotopes on the primary damage production. These calculations result from the integration of neutron-isotopes cross sections with the neutron spectrum of the HFIR facility. All these calculations were performed using the JENDL-4.0 library.

### 3.1. Detailed study of nuclear reaction in the $^{59}\text{Ni}$ isotope

In order to investigate the origin of the two peaks observed on the  $^{59}\text{Ni}$  PKA spectra plotted on Figure 2, PKA spectra associated with the elastic collision, the  $(n, \alpha)$  and the  $(n, p)$  nuclear reactions were plotted on Figure 3. The analysis of these graphs clearly points out that peaks observed on the  $^{59}\text{Ni}$  PKA spectrum are associated with the recoil of  $^{56}\text{Fe}$  and  $^{59}\text{Co}$  and their intensities and positions are function of the incident neutron flux.

In order to estimate the impact of the recoil on the formation of primary damage production, the displacement cross section computed within the BCA framework was computed for different nuclear reactions for the HFIR facility in  $^{59}\text{Ni}$  isotope. Figure 4 displays the variation of the displacement cross section for each nuclear reaction as a function of the kinetic energy of the incident neutron in the laboratory frame.

The analysis of Figure 4 clearly points out that displacement cross section induced by the  $^{56}\text{Fe}$  and  $^{59}\text{Co}$  recoil are dominant over a large energy range. These displacement cross sections are one order of magnitude larger than displacement cross section for the  $^{58}\text{Ni}$  isotope. From the computation of the partial displacement cross section we can conclude that the main part of primary damage produced in Ni based materials results from the recoil atoms induced by nuclear reaction on the  $^{59}\text{Ni}$  isotope. This analysis illustrates the need to

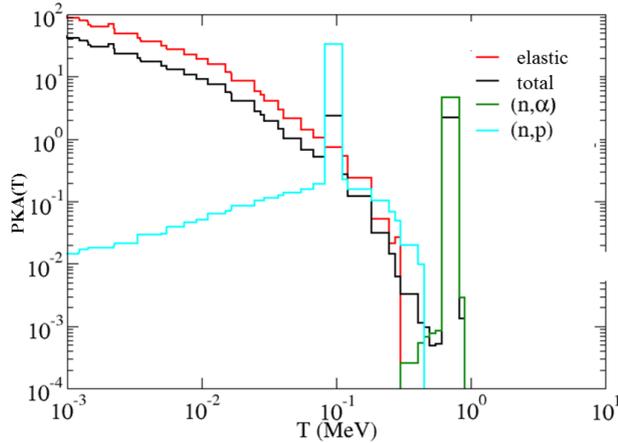


Figure 3: Comparison of PKA spectra as a function of the recoil energy  $T$  for different nuclear reactions occurring in the impinging of the  $^{59}\text{Ni}$  isotope with a neutron (green line :  $(n, \alpha)$  nuclear reaction; blue line:  $(n, p)$  nuclear reaction; red line: elastic collision; black line: all processes are taken into account). All these calculations were performed for the HFIR reactor with the TENDL-2015 library on pure  $^{59}\text{Ni}$  isotope.

compute isotope by isotope the primary damage in materials in order to accurately account all possible nuclear reactions.

### 3.2. Aging of Ni based materials for different facilities

Despite the displacement cross section for  $^{59}\text{Ni}$  isotope is one order of magnitude larger than the one for  $^{58}\text{Ni}$  isotope as pointed on Figure 4, the primary damage production rate (dpa/s) calculated within the BCA framework results from the integration of these displacements over the different neutron fluxes for a given nuclear facility.

To study in detail the effect of aging of material submitted to long time neutron flux, primary damage production rates were computed on 10% Ni based Fe steel for different facilities. To mimic the impact of aging on these damage rates, the primary damage production rates were computed for a pristine sample made of 90% of iron and 10% of  $^{58}\text{Ni}$  isotopes and another sample made of 90% of iron and 10% of  $^{59}\text{Ni}$  assuming that all  $^{58}\text{Ni}$  isotopes are transmuted in  $^{59}\text{Ni}$ .

Figure 5 displays the ratio between the primary damage production rates for different facilities. The analysis of this figure points out that the ratio of primary damage production rates is highly sensitive to the thermal neutron fluxes in agreement with the narrow peaks observed on the displacement cross section of the  $^{59}\text{Ni}$  isotope (cf Figure 4).

Despite the appearance of the  $^{59}\text{Ni}$  isotope is responsible for a large increasing of the primary damage pro-

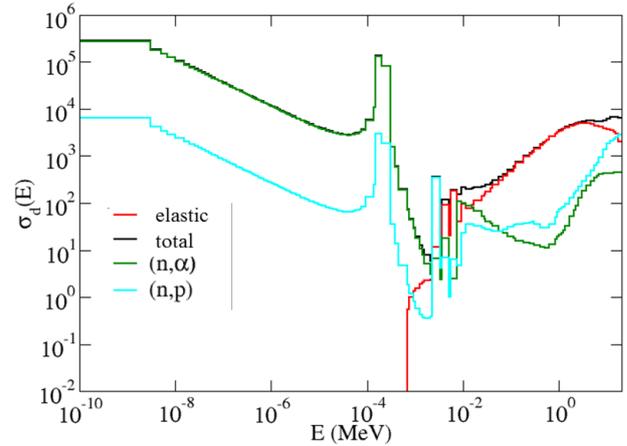


Figure 4: Comparison of the displacement cross sections computed within the BCA framework as a function of the kinetic energy of the incident neutron in the laboratory frame for all nuclear reactions occurring in pure  $^{59}\text{Ni}$  isotope (green line :  $(n, \alpha)$  nuclear reaction; blue line:  $(n, p)$  nuclear reaction; red line: elastic collision; black line: all processes are taken into account). All these calculation were performed for the HFIR reactor with the TENDL-2015 library.

duction rate, this rate is mainly controlled by the fraction of thermal neutrons. For HFIR and PWR facilities exhibiting important thermal neutron fluxes (cf Figure 1), this ratio is important. For 800 keV Ar and 100 keV Xe irradiation, this ratio does not evolve because these irradiation do not induce any nuclear reaction in the material. Fusion facilities like ITER and IFMIF exhibiting small thermal neutron fluxes as displayed in Figure 1 are insensitive to the isotopic composition of the steel.

## 4. Conclusion

To discuss the importance of the isotopic composition on the primary damage, we study in detail the impact of different neutron isotope cross section in Ni elements, composed of different isotopes. From a careful analysis of inelastic neutron isotope cross section using the TENDL-2015 library, it appears that the main part of PKA and displacement cross sections results from the transmutation of the  $^{59}\text{Ni}$  isotope. The analysis of the displacement cross section, calculated by the DART code which computes PKA and displacement cross section for each isotope, exhibit sharp peaks due to the recoils of  $^{56}\text{Fe}$  and  $^{59}\text{Co}$  atoms resulting from the transmutation of the  $^{59}\text{Ni}$  isotope. Calculations performed with the TENDL-2015 library clearly show that these sharp peaks are centered in the epi-thermal domain, i.e. neutron with kinetic energies varying from few hundredth eV to few eV. From this analysis, it was possible

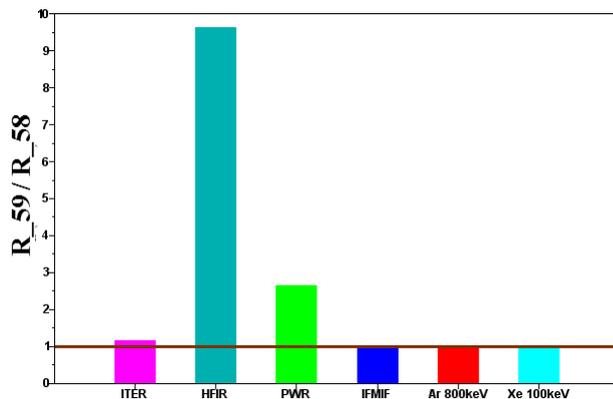


Figure 5: Comparison of the ratio of the primary damage production rate in 10%  $^{59}\text{Ni}$  and 10%  $^{58}\text{Ni}$  based Fe steel (10%Ni and 90% Fe in atomic fraction) for different nuclear facilities. Depending on the neutron spectra of each facility, this ratio can differ from one order of magnitude (red line: ratio equal to unity).

to study the effect of aging in Ni based steels. Despite the transmutation of the  $^{59}\text{Ni}$  isotope is mainly responsible for the high value of the displacement cross section. It appears that only nuclear facilities exhibiting a large amount of thermal neutrons like HFIR facility are sensitive to the aging of Ni based steel, responsible for the vanishing of  $^{58}\text{Ni}$  isotopes and the formation of  $^{59}\text{Ni}$  isotopes.

The example studied in this work clearly demonstrates the need to compute primary damage production from the knowledge of the distribution of isotopes in the material and from very accurate neutron-isotope cross section data libraries.

## Acknowledgement

We acknowledge Dr. Greenwood for providing us the characteristic neutron fluxes of different nuclear facilities, Dr J.-C. Sublet for giving us the TENDL-2015 library and for his helpful advices on the underlying physics beyond the description of inelastic cross sections neutron-isotopes and all members of the international DPA CRP from IAEA for helpful discussions on primary damage calculations.

## References

- [1] L. Luneville, D. Simeone, C. Jouanne, Calculation of radiation damage induced by neutrons in compound materials, *J. Nuclear Materials* 353 (2006) 89–100.
- [2] G. Odette, D. Doiron, Neutron-Energy-Dependent Defect Production Cross Sections for Fission and Fusion Applications, *Nuclear Technology* 29 (1976) 346–368.

- [3] D. Simeone, O. Hablot, V. Micalet, P. Bellon, Y. Serruys, Contribution of recoil atoms to irradiation damage in absorber materials, *J. Nuclear Materials* 246 (1998) 206.
- [4] L. Greenwood, R. Smither, SPECTER: neutron damage calculation for materials irradiations, Tech. Rep. ANL/FPP/TM-187, Argonne National Laboratory (1985).
- [5] M. Griffiths, The effect of irradiation on Ni-containing components in Candu reactor cores: a review, *AECL Nuclear review* 2 (2013) 1–17.
- [6] A. Rowcliffe, L. Mansur, D. Hoelzer, R. Nanstad, Perspectives on radiation effects in nickel-base alloys for applications in advanced reactors, *Journal of Nuclear Materials* 392 (2) (2009) 341 – 352.
- [7] L. R. Greenwood, A new calculation of thermal neutron damage and helium production in nickel, *J. Nuclear Materials* 115 (1983) 137–142.
- [8] A.J. Koning, S. Hilaire and M.C. Duijvestijn, TALYS-1.0, Proceedings of the International Conference on Nuclear Data for Science and Technology, *EDP Sciences* (2008) 211–214.
- [9] R. MacFarlane, D. Muir, The NJOY nuclear data processing system, Tech. Rep. LA-12740-M, Los Alamos (1999).
- [10] T. V. Blosser, J. G. E. Thomas, Neutron Flux and Neutron and Gamma-Ray Spectra Measurements at the HFIR, Tech. Rep. ORNL-TM-2221, Oak Ridge National Laboratory (1968).