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Institut für Neutronenphysik und Reaktortechnik
Projekt Schneller Brüter

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Importance and Status of (n,α)-Cross Sections for a Reliable Prediction of Radiation Damage in Stainless Steel

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Gesellschaft für Kernforschung mbH., Karlsruhe

Abstract

It is well established that helium formed in stainless steel by various (n,α) processes has a pronounced effect on its mechanical and dimensional properties. There has been strong discrepancy between the helium contents measured in irradiated stainless steel and that calculated on the basis of known (n,α)- cross sections on the stable constituents of stainless steel. This discrepancy seems to be resolved by attributing the excessive helium to the two step process: $^{58}Ni(n,\gamma)^{59}Ni(n,\alpha)^{56}Fe$. However, a large discrepancy exists between the value of the thermal cross section for the $^{59}Ni(n,\alpha)^{56}Fe$ process used to calculate the helium production data and that obtained out of the direct measurements of this cross section.

In this paper the role of the (n,α) cross section is investigated on the irradiation induced swelling as an example. It is shown that insoluble gases like helium do influence swelling by way of nucleating and stabilising voids. It is further shown that helium produced via ^{59}Ni constitutes a substantial amount of helium produced in stainless steel under thermal as well as under fast reactor conditions. In fast reactors helium produced via ^{59}Ni gains its importance at high neutron fluences. A linear extrapolation of helium production data measured at low fluences to the fluences of the order of $1.5\cdot10^{23}$ n/cm^2 is not justified. It is shown that the usual practise to relate helium production data to thermal and fast neutron fluence is inadequate. The details of neutron spectrum are necessary to reliably predict helium production rate. A measurement of $^{59}Ni(n,\alpha)^{56}Fe$ cross section in keV and MeV range is recommended.

Bedeutung und Stand der (n, α) Wirkungsquerschnitte für die Voraussagbarkeit von Strahlenschäden in rostfreiem Stahl

Zusammenfassung

Es ist allgemein bekannt, daß das durch verschiedene (n,α)-Prozesse in rostfreiem Stahl produzierte Helium einen deutlichen Einfluß auf dessen mechanische Eigenschaften und Dimensionsänderungen hat. Es wurden große Unterschiede zwischen dem gemessenen Heliumgehalt in bestrahltem rostfreiem Stahl und den anhand der bekannten (n,α)-Wirkungsquerschnitte berechneten Daten beobachtet. Diese Diskrepanz scheint durch die Zuordnung des überschüssigen Heliums zu dem Zwei-Stufen-Prozess $^{58}Ni(n,\gamma)$ $^{59}Ni(n,\alpha)^{56}Fe$ geklärt zu sein. Es existiert jedoch ein großer Unterschied zwischen dem thermischen Wirkungsquerschnitt für den $^{59}Ni(n,\alpha)^{56}Fe$-Prozess, der zur Berechnung der Helium-Erzeugungsrate angewandt wird und dem, der aus direkten Wirkungsquerschnitts-Messungen gewonnen wird.

In diesem Bericht wird die Rolle der (n,α)-Wirkungsquerschnitte als Beispiel am strahleninduzierten Schwellen untersucht. Es wird gezeigt, daß unlösliche Gase wie Helium das Schwellen durch Keimen und Stabilisieren der Poren beeinflussen. Es wird weiter gezeigt, daß sowohl in thermischen als auch in schnellen Reaktoren ein beträchtlicher Teil an Helium über ^{59}Ni gebildet wird. In schnellen Reaktoren gewinnt die Helium-Produktion über ^{59}Ni bei höheren Neutronen-Fluenzen an Bedeutung. Eine lineare Extrapolation der bei niedrigen Fluenzen gemessenen Helium-Produktions-Daten zu Fluenzen von der Größenordnung $1.5 \cdot 10^{23}$ n/cm^2 ist nicht gerechtfertigt. Es wird dargelegt, daß für die Heliumerzeugung das gesamte Neutronenspektrum berücksichtigt werden muß und üblicherweise angewandte Verfahren, bei denen die Heliumerzeugung nur dem thermischen oder dem schnellen Neutronenfluß zugeordnet wird, nicht geeignet sind. Eine Messung des $^{59}Ni(n,\alpha)^{56}Fe$-Wirkungsquerschnitts im keV- und MeV-Bereich wird empfohlen.

CONTENTS

1. Introduction

It is generally recognised that helium formed in stainless steel by various (n,α)-reactions has a pronounced effect on its mechanical and dimensional properties, particularly the high temperature embrittlement, fracture behavior and swelling due to void formation [1-8]. In figure 1 the various categories of radiation damages are schematically shown as a function of temperature and neutron fluence.

Accelerator and reactor experiments have shown that the influence of helium can become significant once its concentration exceeds 1 appm. Bloom [9] has discussed high temperature embrittlement of stainless steel irradiated in fast reactors, concentrating thereby on the synergetic effects of matrix strengthening and helium content. At high temperatures failure occurs by growth and coalescence of helium bubbles on grain boundaries. Although it has been established that the quantity of helium produced in stainless steel cannot account for the observed amount of porosity, helium plays an important role in nucleating, stabilising and subsequent growth of voids. Radiation induced swelling increases with the helium content in stainless steel [5,10].

Although helium in metals can under certain circumstances act independently to degrade the properties of structure materials [11] in general displacement damage and helium content in stainless steel act synergetically to influence the radiation damage properties. Thus utmost care should be taken in the interpretation of experiments in which high helium concentration is attained through α-particle injection to simulate reactor conditions. Since the displacement damage and the helium concentrations typical of fast reactor conditions can not be acchieved simultaneously in an experiment one has to depend on the theoretical prediction. Unfortunately there is no theoretical model available which allows a reliable extrapolation of available data up to a total fast (En>0.1 MeV) neutron fluence of $8\times10^{22}\,n/cm^2$ to the fluence typical for the fast reactors ($15-25\times10^{22}\,n/cm^2$). Possibly the lack of exact experimental data is the reason for the difficulty in establishing a profound theoretical model.

Major helium producing constituents of stainless steel are Fe, Cr, Ni and in some steels also B and N. The cross sections for these materials are stored on different nuclear data libraries like (KEDAK, UKNDL and ENDF/B).

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The evaluations are based on existing experimental and theoretical information. Though the data in different libraries differ it is believed that on the whole they are not drastically wrong. Until 1971 there was a strong discrepancy between the helium generation rate calculated on the basis of known (n,α) cross sections and measured in reactor experiments [12]. To resolve this discrepancy a two step process was presumed to cause the excessive generation of helium. Bauer and Kangilaski [13] accounted the excessive helium to the two step process

$$^{58}Ni(n,\gamma)^{59}Ni(n,\alpha)^{56}Fe.$$

The thermal cross section for the first process being about 4 barn [14]. Bauer and Kangilaski estimated that a thermal cross section of 7.39 barn for the $^{59}Ni(n,\alpha)^{56}Fe$ process would explain the excessive generation of helium in stainless steel and nickel. This value, as will be shown later, differs considerably from the value obtained out of the direct measurements of this cross section.

An absence of a profound theoretical model renders it difficult to make direct quantitative correlations of different radiation damages to the gas production cross sections. In spite of this difficulty, we will take radiation induced swelling as an example to demonstrate qualitatively the influence of gas production on the radiation damage.

In the next chapter the importance and implications of swelling on reactor design are summarised and in the third chapter some of the major observations of the experimental data regarding void formation are given. In chapter 4 the mechanism of void formation is studied. Chapter 5 deals with the available (n,α)-cross sections. The last chapter contains recommendations for future work.

2. Impact of swelling on reactor components

Positive, reliable and reproducible fixation of the fuel relative to the reactors geometric centre is highly important because of the neutronic sensitivity of fast reactors to dimensional changes. The reactivity of a typical 1000 MW fast reactor changes with the dimensional changes of the reactors as follows [15]:

$$\frac{\Delta k}{k} \sim - 0.5 \frac{\Delta r}{r}$$

and

$$\frac{\Delta k}{k} \sim - 0.3 \frac{\Delta h}{h}$$

i.e. an increase in core radius by 0.7 % or hight by 1.2 % reduces the reactivity by $1\ \$$. ($\frac{\Delta k}{k}$ = 0.0035)

The core materials may be divided according to their stay in the reactor in two categories

1. Fuel assemblies
2. Core structure

The fuel assembly has a short service life of about 3 years in a reactor. Fuel assemblies are expendable and as such have no inherent design permanence. The plant owner can capitalise any scientific break through and advances in the material science during the life time of the reactor.

Although the stainless steel swelling was first observed on fuel element claddings in the U.K. fast reactor program [13] core designers do not consider clad-swelling as a major fuel element problem. Some investigators [16,17] even regard this phenomenon as a potential promoter of higher burnup rather than cladding failure.

The core structure and blanket implicate the hardware of the reactor which is permanently installed for the lifetime of the reactor which is of the order of 30 years. During this period the materials are exposed to high neutron fluence and temperatures from 350°C to 600°C. Allowances have to be made in reactor design to accommodate swelling under these conditions.

For the design conception of a fast breeder reactor in Germany an allowance of 10 % swelling is accommodated. As is shown in Figure 2 (taken from ref. 18) for SNR 2 the swelling will be under 10 % only if the basic theory and data of the optimistic model used for extrapolation of the existing data are confirmed to be valid. In the following chapter the important observations of the available experiments are summarised.

3. Experimental observations on void formation

Since the detection of voids and dislocations during fast neutron irradiation by Cawthorne and Fulton in 1966 this subject has been widely studied in different experiments. The important qualitative observations are

1. There appears to be a fluence threshold above which voids are observed.

2. This threshold increases with increasing temperature.

3. At low void concentrations a significant fraction of void concentration is associated with dislocations or precipitate particles. At higher concentrations these locations are difficult to detect.

4. Although the void concentration and size distribution reported by different authors vary considerably the bulk volume changes of the material are similar.

5. The composition of the stainless steel is an important variable influencing void concentration and swelling.

6. Helium produced in stainless steel by various (n,α) reactions can severely limit the ductility of steel at elevated temperature.

7. The helium content enhances void formation and swelling significantly. The concentration of voids and their size depend critically on helium content.

8. Although most of the radiation damage effects are synergetical effects of displacement damage and helium production, the latter can also act independently to degrade the property of reactor structure components.

9. Protons produced by various (n,p) reactions do not have a measurable influence on the properties of the materials. This may be attributed to the high solubility and high mobility of protons [5].

In order to have a better understanding of the role of the (n,α) process a possible mechanism which may be responsible for the enhancement of swelling due to helium is investigated.

4. Mechanism of helium induced swelling

It has been mentioned that swelling is a synergetic effect of helium production and displacement damage. In this chapter it is outlined how the helium produced by irradiation effects the void formation and enhence the swelling at high temperatures.

The irradiations either with neutrons or with electrons or ions have the effect that vacancies and interstitials are produced in the metals with concentrations well in access of their thermal concentrations.
A large aggregate of vacancies is called void. The classical theory can not predict the rate of formation of voids because of simultaneous existence of excess vacancies and excess interstitials. In 1971 Katz and Wiedersich [19] and Russel [20] have developed theories to calculate formation of voids when both vacancies and interstitials are supersaturated.

In the following the mechanism of the formation and growth of voids is outlined without going into mathematical details.

If only the vacancies are supersaturated, the nucleation rate of voids is determined by the random walk over a free energy barrier. Vacancies can collide together to form a multiple vacancy. A single vacancy may form a di-vacancy and a di-vacancy may emit a vacancy and thus again form two single vacancies or may capture a vacancy to form a trivacancy. In general, a n-vacancy cluster may emit or capture a single vacancy to form a (n-1) or (n+1) vacancy cluster.

As long as the cluster is smaller than the critical size corresponding to the maximum free energy, the vacancy emission process is more probable than the vacancy capture. Clusters larger than the critical size are more likely to grow than to shrink. Thus many clusters start to grow but evaporate again before the critical size is reached.

The mobile interstitials which are produced together with vacancies during irradiation introduce another process, namely interstitials capture which causes vacancy clusters to shrink from n-vacancy clusters to (n-1) vacancies. The effective energy barrier is broadened (dashed line in figure 3).

Helium (or other insoluble gases) produced during irradiation, has a stabilising effect on the vacancy clusters. Because of the long range of neutrons helium is produced throughout the structural material and in direct vicinity of the vacancies. This helium can be captured into a vacancy cluster to form gas containing voids. The vacancies are less likely to be emitted from a gas containing void than from a gas free void. This can be explained as follows:

The frequency of vacancy emission is determined by the work required to emit a vacancy (i.e. free energy change of the void when a vacancy is emitted). A decrease in the void size of a n-vacancy cluster containing helium atoms would require the gas pressure inside the void to increase. That means *pv* work must be done to compress the gas. Hence the presence of gas in the void would decrease the vacancy emission rate.

The vacancy or interstitial capture rate is determined by the vacancy or interstitial flux. Their capture rate is, therefore, not effected by the gas present in the void.

The presence of helium thus has two effects on vacancy cluster shrinkage. It decreases the vacancy emission rate because of the increase in the free energy required for this process and it blocks the void shrinkage below a certain minimum size required to accommodate helium atoms.

Both effects increase the void nucleation rate and depend upon the magnitude of helium flux. If helium flux is comparable to vacancy flux, clusters will acquire helium atoms as fast as they acquire vacancies and vacancy emission would never occur from a void. In this case helium will drive the nucleation process and the nucleation rate will be limited by the arrival rate of the vacancies at the helium filled clusters.

In addition to the capture of helium at vacancy clusters helium can also condense on the grain boundaries and dislocations. This process besides increasing the swelling also contributes significantly to the high temperature embrittlement of stainless steel.

It is evident that though helium can not account for the total swelling of stainless steel it vitally influences this process. Therefore for a reliable prediction of voids or swelling, vacancy and helium flux rate should be known with sufficient accuracy and care should be taken in extrapolating the results of implantation experiments, if the gas flux and vacancy flux conditions are very much different from those encountered in reactors.

5. Status of gas production cross sections

As has already been remarked the (n,p) process, due to the high solubility and high mobility of hydrogen in metals, has little effect on the behavior of stainless steel under irradiation. Therefore this chapter will be confined to the helium production data only.

After it was first established in 1967 that helium can play an important role in downgrading the properties of reactor structural materials, say stainless steel, a number of experiments have been performed to measure the helium production rate in stainless steel. In metal physics it is customary to relate the helium production to thermal and fast neutron fluences. For thermal neutron fluences ranging from about 3×10^{22} to 10^{23} n/cm^2 the helium production is reported to follow the following proportionality [5)]

$$C(He) \sim (\phi t)^{1.68}$$

where ϕt is the equivalent thermal neutron fluence. It is seen in table 1 that all the consituents of stainless steel show a threshold behaviour with exception of ^{10}B. But the $^{10}B(n,\alpha)$cross section which is well known can not account for the amount of helium produced in stainless steel. Moreover the helium produced by any direct (n,α) process would not account for the exponent in the above equation rather some two step process should be considered to account for the departure from linearity in equation 1. The two step process first proposed was [21,22)]

$$^{58}Ni(n,p)^{58}Co(n,\alpha)^{55}Mn.$$

Table 1: (n,α)-reaction data for constituents of stainless steel

Isotope	Q-value [MeV]	Excitation Energy of Compound nucleus [MeV]	Threshold (KEDAK-3) [MeV] ≈	<σ> [mb] SNR-2-Spectrum	<σ> [mb] Fission spectrum
^{50}Cr	0.3	9.26	4.0	0.084	1.49
^{52}Cr	-1.2	7.98	5.0	0.0114	0.215
^{53}Cr	+1.8	9.72	4.0	0.018	0.313
^{54}Cr	-1.55	6.28	7.0	0.0007	0.015
^{54}Fe	0.848	9.3	4.0	0.058	1.01
^{56}Fe	0.322	7.65	5.0	0.0082	0.164
^{57}Fe	2.402	10.04	5.0	0.005	0.103
^{58}Fe	-1.387	6.59	7.0	0.0013	0.029
^{58}Ni	2.89	9.0	2.0	0.419	6.16
^{59}Ni	5.094	11.39	-	72.92	33.2
^{60}Ni	1.35	7.8	4.15	0.046	0.80
^{61}Ni	3.57	10.6	6.0	0.0086	0.18
^{62}Ni	-0.44	6.84	8.0	0.00018	0.004
^{64}Ni	-2.43	6.1	12.75	0.0	0.0
^{92}Mo	3.69	8.1	7.85	0.0031	0.074
^{94}Mo	5.13	7.4	6.0	0.019	0.394
^{95}Mo	6.4	9.2	4.8	0.015	0.283
^{96}Mo	4.0	6.8	6.5	0.0036	0.078
^{97}Mo	5.37	8.6	6.2	0.00077	0.016
^{98}Mo	3.2	5.9	8.0	0.00046	0.011
^{100}Mo	2.4	5.4	8.56	0.0001	0.0023
^{31}P	-1.94	7.94	4.0	0.1046	1.84
^{14}N	-0.16	10.8	1.0	7.56	85.85
^{12}C	-5.7	5.0	7.2	0.053	0.55
^{10}B	+2.8	11.5	-	3302.0	0.44

If this process is to account for the excessive helium production in stainless steel, the cross section for the first reaction being very small ≤ 70 mb (ref. 23), a high cross section of about 1000 b is required for the $^{58}Co(n,\alpha)^{55}Mn$ process. As has been shown earlier [12] this value is not a probable one. Thus this process has to be discarded as a source of excessive helium production. In subsequent experiments nickel was confirmed to be a source of enhanced helium production [13,24].

It is now well accepted [5,7] that the two step process responsible for excessive helium production in stainless steel is

$$^{58}Ni(n,\gamma)^{59}Ni(n,\alpha)^{56}Fe.$$

^{59}Ni has a half life of 7.5×10^4 years. The cross section for the $^{59}Ni(n,\alpha)$-process derived from the helium production data ranges from 7.38 b to 13 b for thermal neutrons. In the material research community the lower value of 7.38 b derived by Bauer and Kangilaski [13] is widely used and good agreement between the calculated and measured helium production data is reported. It is even concluded that cross sections for (n,α) processes are well known [25].

In recent years a number of direct measurements for the $^{59}Ni(n,\alpha)^{56}Fe$ cross section have been reported for thermal neutrons. The results are reproduced in table 2.

Table 2: $^{59}Ni(n,\alpha)^{56}Fe$ cross section for thermal neutrons reported from different laboratories

Laboratory	year	cross section
Knoll Atomics[26]	1974	13.7 ± 1.2 b
Chalk River [27]	1974	18.0 ± 1.6 b
Chalmer Univ.[28] Göteborg	1975	22.3 ± 1.6 b
ORNL[29]	1975	12.0 ± 1 b

It is seen that even if the lowest value of these direct measurements is used an average over the Maxwell spectrum would lead to a value of 10.64b. This value is about 50 % higher than the value derived by Bauer and Kangilaski and widely used in material research.

The reason for the afore mentioned agreement between the calculated and measured helium production data for stainless steel in spite of the low value used for the $^{59}Ni(n,\alpha)$ cross section may be; a) the helium production is not sensitive to the $^{59}Ni(n,\alpha)$ cross section. This argument can not be maintained in the light of what has been said before. b) The accuracy of the helium production measurements is so low that no conclusions can be drawn out of these experiment on the (n,α) cross sections. c) The inherent similarity of different experiments introduces some systematic errors in the experimental determination of helium production that an apparent agreement is duped with the low value of thermal $^{59}Ni(n,\alpha)$ cross section. May be some of the helium produced during irradiation escapes detection say by diffusing out of the sample.

To assess the error made by using the low value of 7.38 b for the thermal $^{59}Ni(n,\alpha)^{56}Fe$ cross section. The data of reference 25 are recalculated using the lowest value out of the direct measurements for this cross section i.e. the thermal cross section for the $^{59}Ni(n,\alpha)^{56}Fe$ process is taken to be 10.64 b. For the boron-free sample the present calculation gives about 40 % higher helium production rates than calculated in reference 25. This means that for one lower-fluence data point the agreement between the calculated and measured helium production rate is improved and for other data points a deviation between 25-40 % is obtained. In the boron-containing stainless steel at low fluence values the helium production via the $^{10}B(n,\alpha)^{7}Li$ reaction dominates and the effect of the two step process becomes only eminent once fluence values higher than 10^{21} n/cm^2 are reached, that is after all the boron has been consumed. While considering the figures reported above it should be kept in mind that the value for $^{59}Ni(n,\alpha)^{56}Fe$ cross section used in this calculation is the lowest value obtained out of direct experiments. An evaluated value, considering all the measurements, would lead to a much higher value for the $^{59}Ni(n,\alpha)^{56}Fe$ cross section. Moreover since the details of neutron spectrum are not available, any contribution to the helium production via the two step process $^{58}Ni(n,\gamma)$ $^{59}Ni(n,\alpha)^{56}Fe$ for non-thermal neutrons could not be included in the present calculation. An inclusion of this contribution, as will be seen later, will further enhance the helium production rate.

After ^{10}B with a large thermal cross section for the (n,α) process of 3836 b has been burned up, no significant source of the (n,α) reaction other than on ^{59}Ni is known for thermal neutrons. At higher neutron energies, of importance in fast reactors, (n,α) reactions on the stable isotopes constituting stainless steel also contribute to the helium production. In table 3 one group cross sections weighted with the spectrum of a proposed fast reactor (SNR-2) are given for the major constituents of stainless steel.

Table 3: One group (n,α) cross section for the major constituents of stainless steel (type 316) weighted with the proposed SNR-2 spectrum

Element	Abundancy	Atom/gram SS	$\langle\sigma\rangle$ mb	$N \cdot \langle\sigma\rangle \left[\frac{cm^2}{gram\ SS}\right]$
Fe	63.6 %	6.814×10^{21}	0.011	7.5×10^{-8}
Cr	18 %	2.077×10^{21}	0.015	3.1×10^{-8}
Ni	13 %	1.345×10^{21}	0.3	4.0×10^{-7}
Mo	2.6 %	0.163×10^{21}	0.0054	8.8×10^{-10}
Mn	1.9 %	0.207×10^{21}	0.0048	1.0×10^{-9}
				Total 5×10^{-7}

The (n,α)-process on stable isotopes of Cr, Fe, Ni and Mo show a threshold behavior even in those cases where the process is exothermic. This is seen in table 1 and figure 4. The reason for this threshold behavior is that the high coulomb barrier of about 10 MeV inhibits the emission of α-particles from the compound nucleus. In the case of ^{59}Ni, however, the exitation energy of the compound nucleus is high enough to overcome the coulomb barrier even at low neutron energies. The $^{59}Ni(n,\alpha)^{56}Fe$ cross section is derived from whatever scarce information available. The data for the 203 eV resonance are rather well measured and the cross section at lower energies is mainly determined by this resonance. Above this resonance 15 more resonances have been observed by Harvey et al. [29] but the parameters for these resonances are not completely established. Above 20 keV the data shown in the upper part of figure 5 for the $^{59}Ni(n,\alpha)^{56}Fe$ process correspond to the calculations of Kirouac. The $^{58}Ni(n,\gamma)$ cross sections are taken from KEDAK-3 [12]. The one group cross sections obtained with the SNR-2 spectrum for the process $^{58}Ni(n,\gamma)^{59}Ni(n,\alpha)^{56}Fe$ are:

for $^{58}Ni(n,\gamma)^{59}Ni$ = 22.2mb and

for $^{59}Ni(n,\alpha)^{56}Fe$ = 50.8mb.

With these data the helium production in a central zone of the SNR-2 reactor is calculated and compared with the helium production with the (n,α) process on the constituents of stainless steel. It is seen in figure 6 that after the end of the first eighteen months, i.e. when the fuel assembly has absolved one half of its residence time in the reactor, 20 % of the helium is produced via the $^{58}Ni(n,\gamma)^{59}Ni(n,\alpha)^{56}Fe$ process. When the fuel assembly leaves the reactor after three years with a total neutron exposure of about 5×10^{23} n/cm^2 the helium produced via ^{59}Ni amounts to about 50 % of the helium produced otherwise.

It is also seen in figure 6 that up to a neutron fluence of less than 8×10^{22} n/cm^2, for which experimental data for the helium production in stainless steel are available, the helium production via the two step process does not come appreciably into affect. The linear dependance of the helium generation rate on fluence observed at low fluences would not hold at high fluence values of the order of 1.5×10^{23} n/cm^2.

To study the sensitivity of helium production on the value of $^{59}Ni(n,\alpha)^{56}Fe$ cross section, its spectral average value is doubled which is well within the limits of the uncertainty of this cross section. The result of the calculation with this doubled value of the (n,α) cross section is shown with broken line in fig. 7. It is seen that if this value of the $^{59}Ni(n,\alpha)$ cross section is used and the fuel assembly is allowed to stay in the reactor for three years or more the amount of helium produced via ^{59}Ni dominates over that produced by direct (n,α) reaction on the stable constituents of stainless steel.

The structural material at different positions of reactors will be exposed to neutrons with different energy spectra. The mean energy of the neutron decreases towards the outer core regions and the blanket. It is expected that the helium production rate will depend upon the location of the material within the reactor. To investigate the spatial dependance of the helium generation one group cross sections have been calculated with different weighting spectra (Fig. 8) corresponding to each of the five zones of an international LMFBR-benchmark reactor [30].

It is seen in table 4 that <σ> varies strongly from one zone to the other i.e. it shows strong spectral effects.

Table 4: One group cross sections for the $^{59}Ni(n,\alpha)^{56}Fe$ process for different reactor zones of an international LMFBR-Benchmark

zone	<σ>
inner core	29.32 mb
outer core	19.71 mb
Axial Blanket	210.2 mb
Radial Blanket	142.0 mb
Reflector	1234.1 mb

To study the flux effect on helium production in stainless steel the neutron flux is usually correlated to thermal neutron flux and fast neutron flux with neutron energies above 0.1 MeV or 1 MeV on occasions even above 3.68 MeV. The neutron energies from thermal to 100 keV are not taken into account in this classification. This classification is justified only if the helium production through the (n,α)-process on stable isotopes contained in stainless steel is considered. As has been shown (Fig. 6) a substantial amount of helium is produced via ^{59}Ni and the $^{59}Ni(n,\alpha)^{56}Fe$ cross section (Fig. 5) shows a resonance structure in the energy range from 100 eV to 20 keV with the peak cross section rising well above 100 b for the 203 eV resonance. Therefore the classification of flux in the above mentioned categories is inadequate to predict the helium production rate in nickel based stainless steel, rather the details of the energy dependence of the flux in eV and keV range should be taken in account.

6. Conclusion

It is seen (chapter 3 and 4) that radiation induced swelling is a synergetic effect of displacement damage (vacancy flux) and helium production rate. Therefore any physically meaningful model to calculate the radiation induced swelling of stainless steel should contain both of these parameters. An adequate knowledge of these parameters is necessary to reliably extrapolate the swelling data to fast reactor conditions.

A substantial amount of the helium production in fast reactor flux takes place via the reactions

$$^{58}Ni(n,\gamma)^{59}Ni(n,\alpha)^{56}Fe$$

Both these reactions exhibit a resonance structure in the keV region. The usual practice to relate helium production data to thermal and fast neutron flux is not adequate to account for the helium production via ^{59}Ni: Specially because fast neutrons in such a classification are defined as neutrons with energy above 100 keV or 1 MeV. As it is seen in Fig. 5 high $^{59}Ni(n,\alpha)$ cross section values are encountered for neutrons with energies less than these limits. The one group cross section for the $^{59}Ni(n,\alpha)$ process (Table 4) is sensitive to flux changes in the eV and keV region. Therefore it is recommended that in future work to calculate the helium production rate in nickel based stainless steel due account of the details of neutron flux should be taken.

The state of the knowledge of the $^{59}Ni(n,\alpha)^{56}Fe$ cross section above 200 eV is very unsatisfactory. Practically no reliable experimental data are available above the 203 eV resonance. Some data up to 20 keV are reported by Harvey [31] at the Lowell Conference but much work is to be done before these data can be finalised. Above 20 keV there is no experiment reported as yet. It is therefore recommended that the $^{59}Ni(n,\alpha)^{56}Fe$ cross section should be measured in the keV and MeV range. An accuracy of about 20 % is considered sufficient for this cross section.

Since nickel is the major contributor to the helium production in stainless steel it will be worthwhile to check whether the assumption made in this work and at other laboratories that the value of (n,α) cross section for the stable nickel isotopes is negligible below 2 MeV is correct or not. Looking at table 1 a possible candidate for this check may be ^{61}Ni with a Q value of 3.57 MeV. For this isotope the thermal cross section of 45 mb has been reported in the past [37] but a recent measurement [28] could not confirm this value and predict the $^{61}Ni(n,\alpha)$ cross section for thermal neutrons to be less than 2 mb. In fact a measurement of the (n,α) cross section in the keV and MeV range on elemental nickel will increase the confidence on helium production calculations. Till date there are practically no measurements of the (n,α) cross section for this important material at energies lower than 14 MeV. The evaluations of the (n,α) cross sections are mostly based on systematic studies.

References

1) A.S. Fraser, I.R. Briss and C.C. Cawthorne,
Nature 211 (1966) 291

2) D. Kramer, H.R. Brager, C.G. Rhodes and A.G. Pard,
J. Nucl. Mater 25 (1968) 121

3) R.S. Nelson and D.I. Mazey,
Radiation Damage in Reactor Materials, Symposium Proceedings,
IAEA Vienna, (1969) Vol. II, p.157 (STI/PUB/230)
and J. Nucl. Mater. 37 (1970) 1

4) H.R. Brager, J.L. Straalsund, J.J. Holmes and J.F. Bates,
WHAN-FR-16 (1970)

5) W.N. McElroy and H. Farrar IV,
Radiation-Induced Voids in Metals, Proceedings of Conference held at Albany N.Y.,
June 9-11, 1971, p. 187

6) M. Bocek and K. Ehrlich,
J. Nucl. Mater. 58 (1975) 247

7) E.E. Bloom and F.E. Wiffen,
J. Nucl. Mater. 58 (1975) 171

8) G. Das and T.E. Mitchell,
J. Nucl. Mater. 56 (1975) 297

9) E.E. Bloom,
Irradiation Embrittlement and Creep in Fuel Cladding and Core components,
British Nuclear Energy Society London (1973) p. 93

10) R.S. Barnes,
J. Nucl. Mater. 11 (1964) 135

11) F.W. Wiffin and E.E. Bloom,
Nucl. Technol. 25 (1975) 113

12) B. Goel,
unpublished (1975)

13) A.A. Bauer and M. Kangilaski,
J. Nucl. Mater. 42 (1972) 91

14) B. Goel,
KFK 2233 (1975)

15) P.R. Huebotter and T.R. Bump,
Radiation-Induced Voids in Metals, Proceedings of the Conference
held at Albany N.Y., June 9-11, 1971

16) C. Cawthorne and E.J. Fulton,
Nature 216 (1967) 575

[17] T.R. Bump,
Nucl. Appl. & Technology 9 (1970) 301

[18] K. Ehrlich and W. Dietz,
Atoomenergie 18 (1976) 189

[19] J.L. Katz and H. Wiedersich,
J. Chem. Phys. 55 (1971) 1414

[20] K.C. Russel,
Acta Met. 19 (1971) 753

[21] J. Weitman and N. Daverhög,
Proc. Conf. Neutron Cross Section and Technology, NBS-special publication 229 (1968) 125

[22] J. Weitman, N. Daverhög and S. Farvalden,
Trans. Am. Nucl. Soc. 13 (1970) 557

[23] W.L. Iijp,
RCN-37 (1965)

[24] I.R. Briss and W.E. Elliss,
Proc. Brit. Nucl. Soc. Conf. Reeding University (1971) 339

[25] K.D. Closs,
Atomwirtschaft 19 (1974) 134

[26] H.M. Eiland and G. Kirouac,
Nucl. Sci. Eng. 53 (1974) 1

[27] R.D. Werner and D.C. Santry,
Nucl. Sci. Eng. 56 (1975) 98

[28] J. McDonald and N.G. Sjöstrand,
Atoomkernenergie 27 (1976) 112

[29] J.A. Harvey, N.W. Hill, J. Halperin, S. Raman and R.L. Macklin,
ORNL 5137 (1976) p.2

[30] C.H.M. Broeders,
Private communications (1977)

[31] J.A. Harvey,
Proc. International Conference on Interaction of Neutrons with Nuclei, Lowell, July 6-9 (1976) p. 143

[32] F. Münnich,
Zeitschr. Physik 153 (1958) 106

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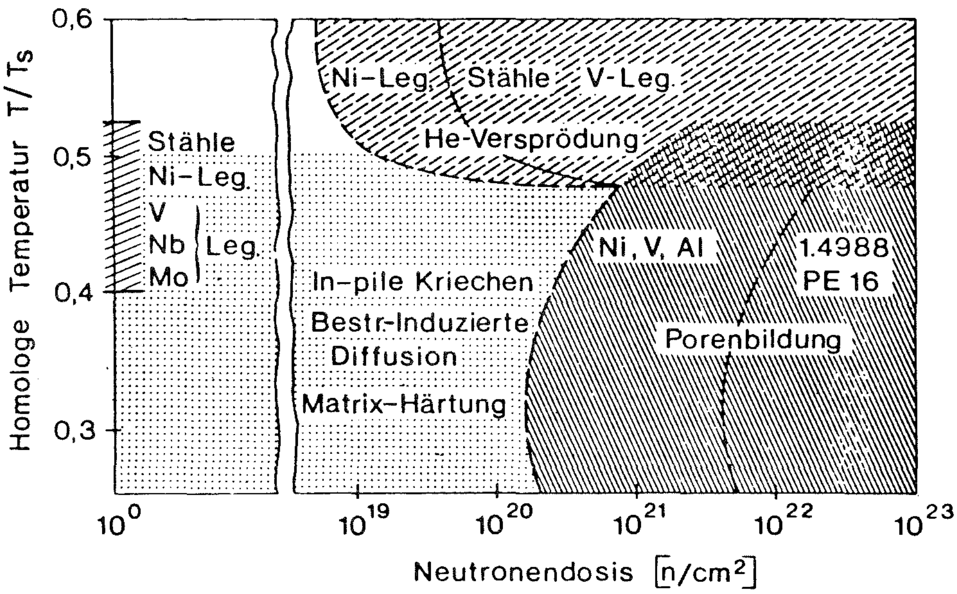
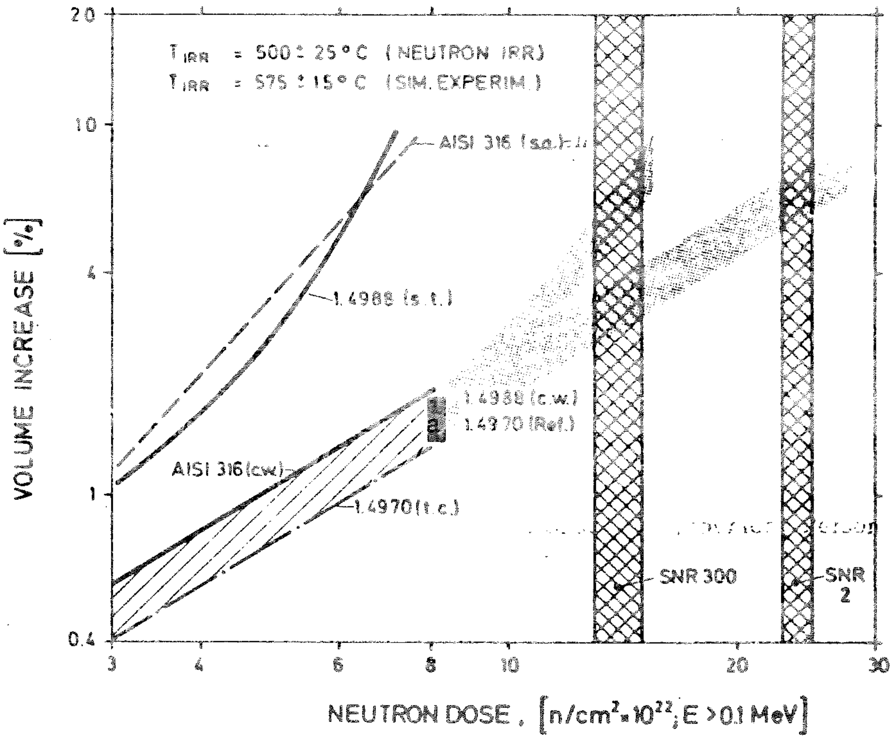


Fig. 1: Important radiation damages as function of neutron dose and temperature



<u>Fig. 2:</u> Possible extrapolation of swelling of stainless steel for neutron dose relevant to SNR-300 and SNR-2 at $500^{\circ}C$ irradiation temperature

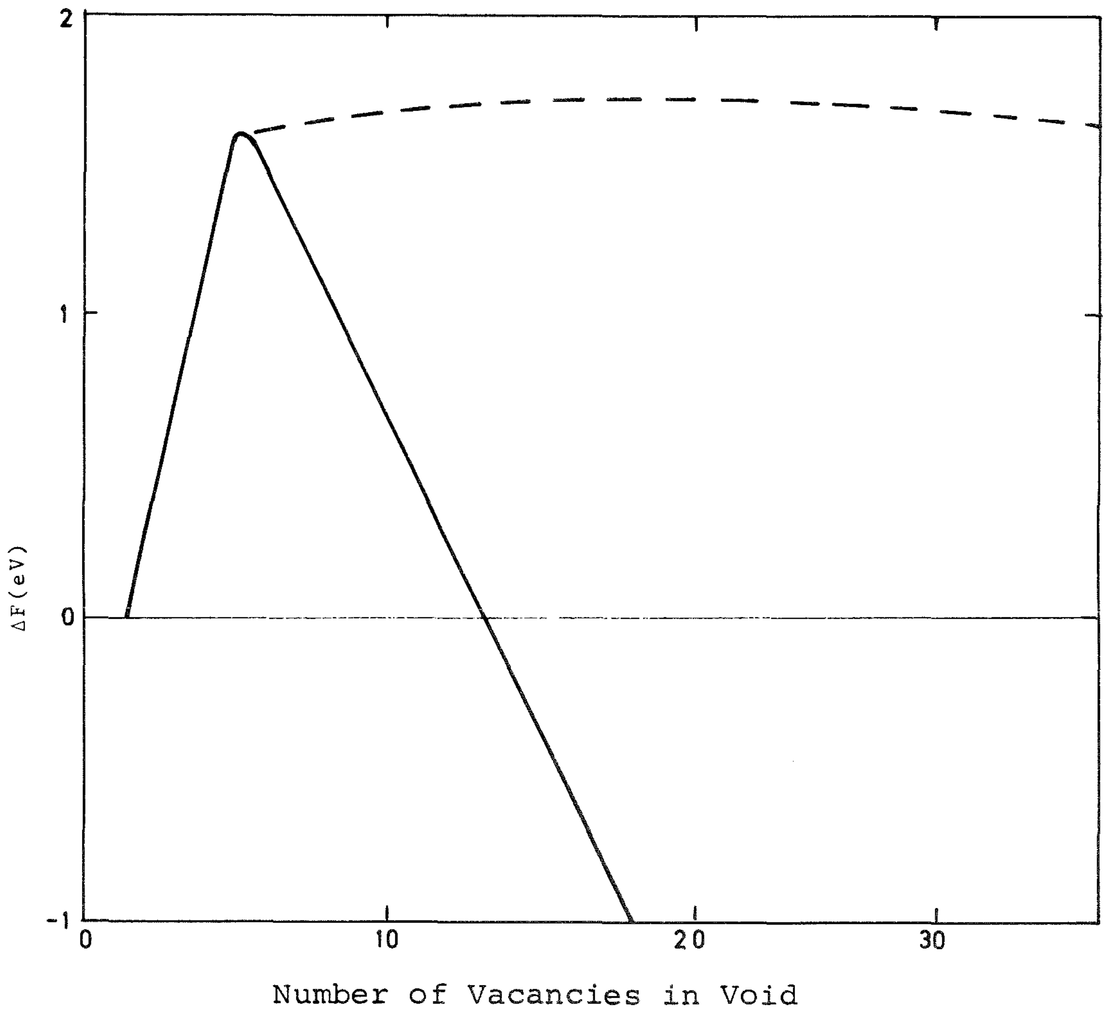


Fig. 3: Free energy barrier for nucleation of voids as a function of a number of vacancies in the cluster. Solid line denotes free Energy barrier in the absence of interstitial dashed line with interstitial flux 0.95 lines the vacancy flux.

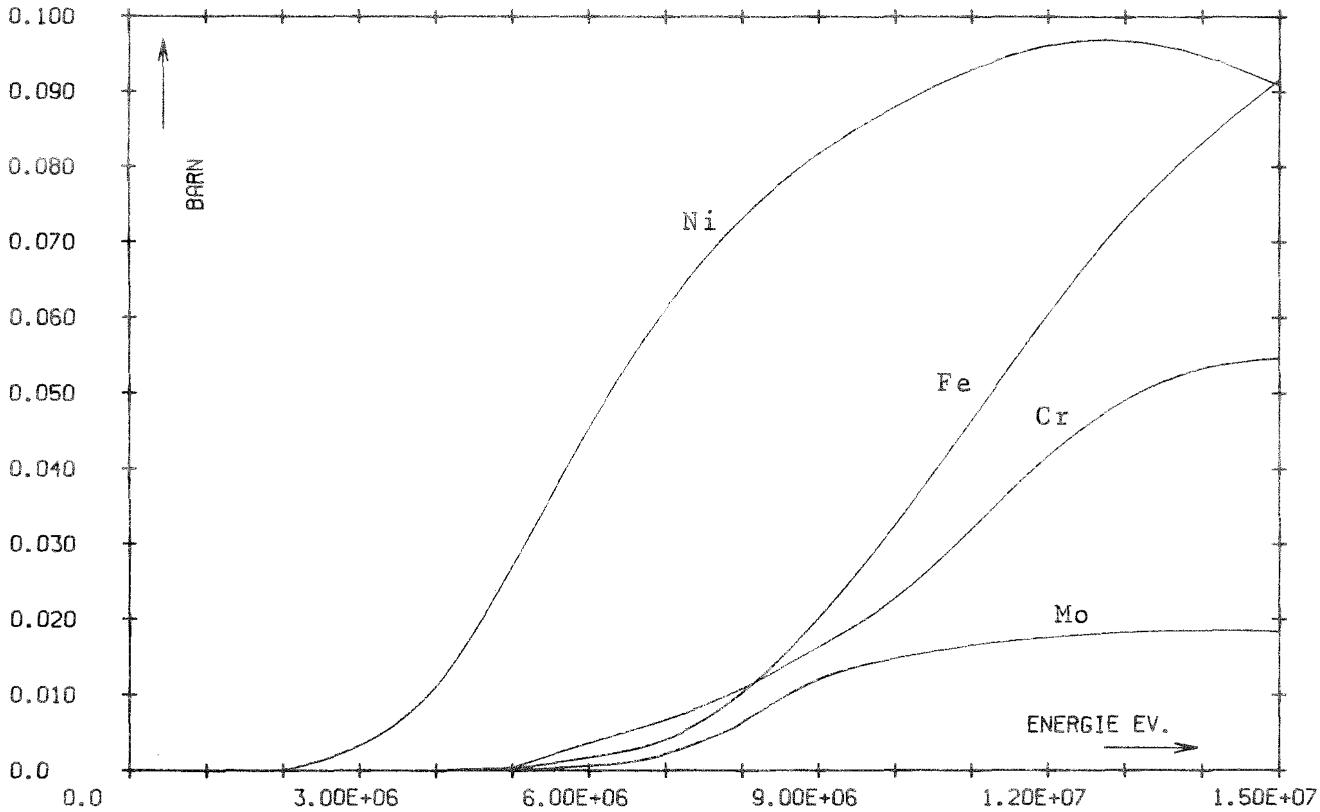


Fig. 4: (n,α) cross sections of the major constituents of stainless steel

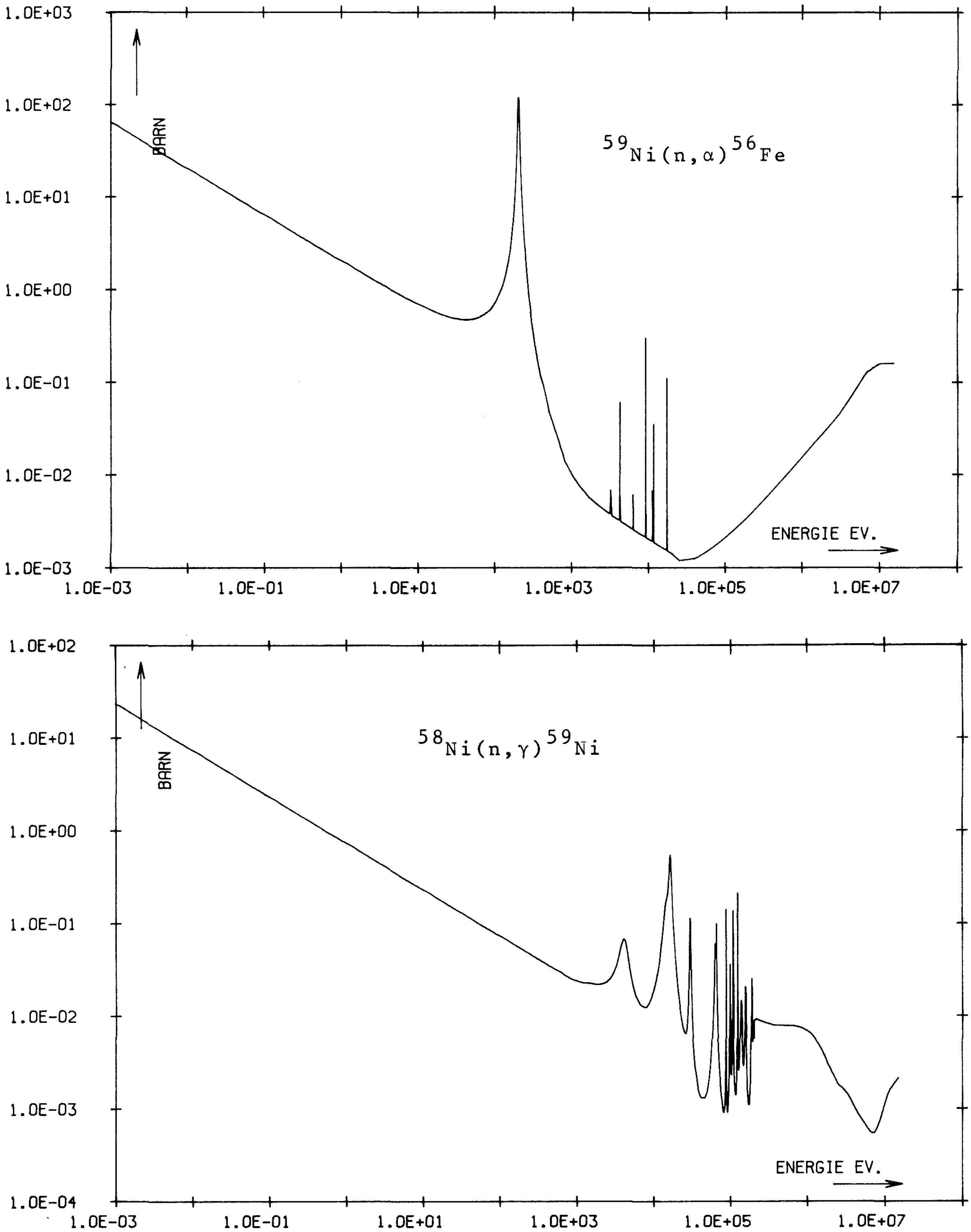


Fig. 5: Cross sections for the process $^{58}Ni(n,\gamma)^{59}Ni(n,\alpha)^{56}Fe$

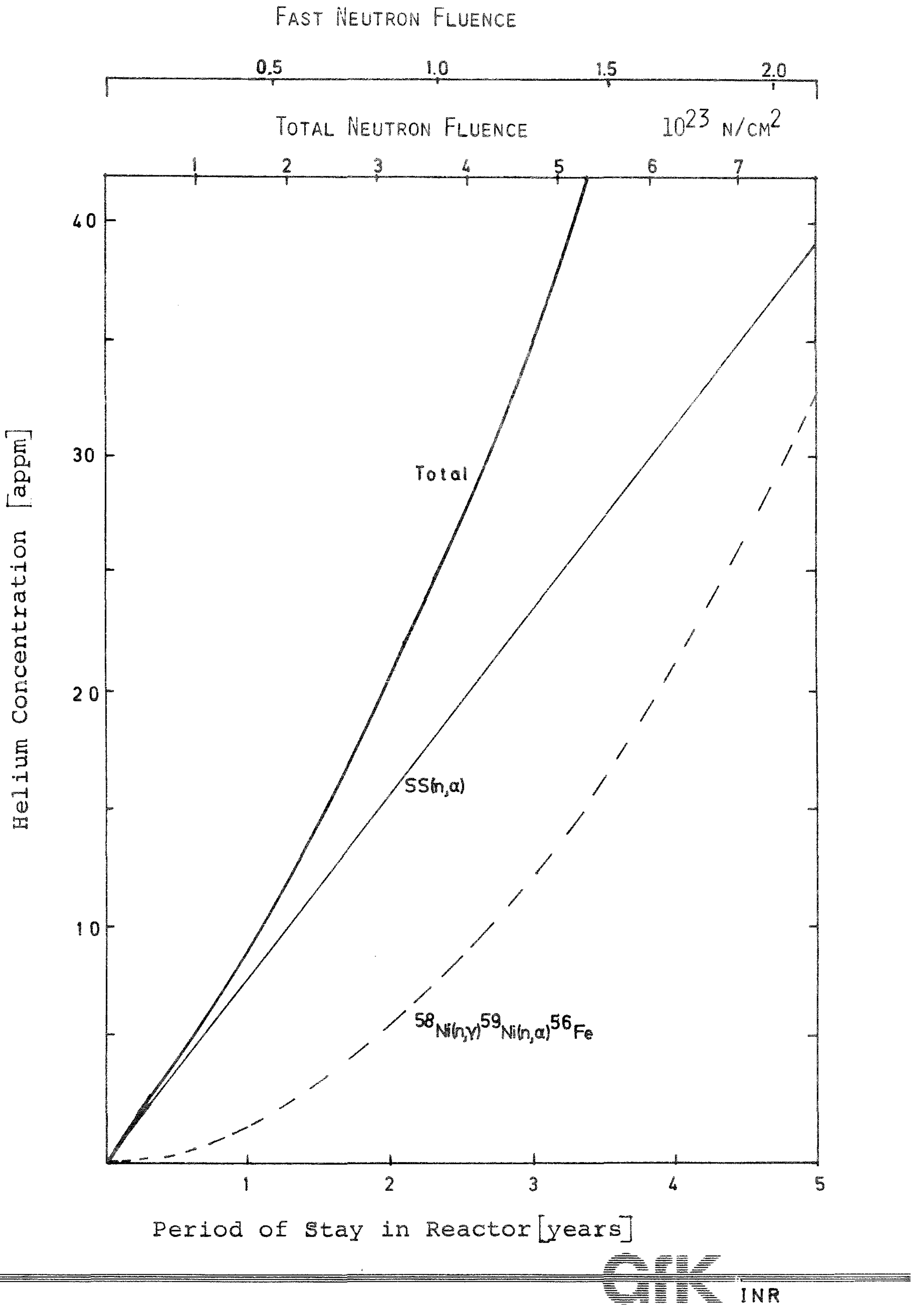


Fig. 6: Helium generation in stainless steel. Thin solid line denotes helium generation in the absence of two step process (dashed line)

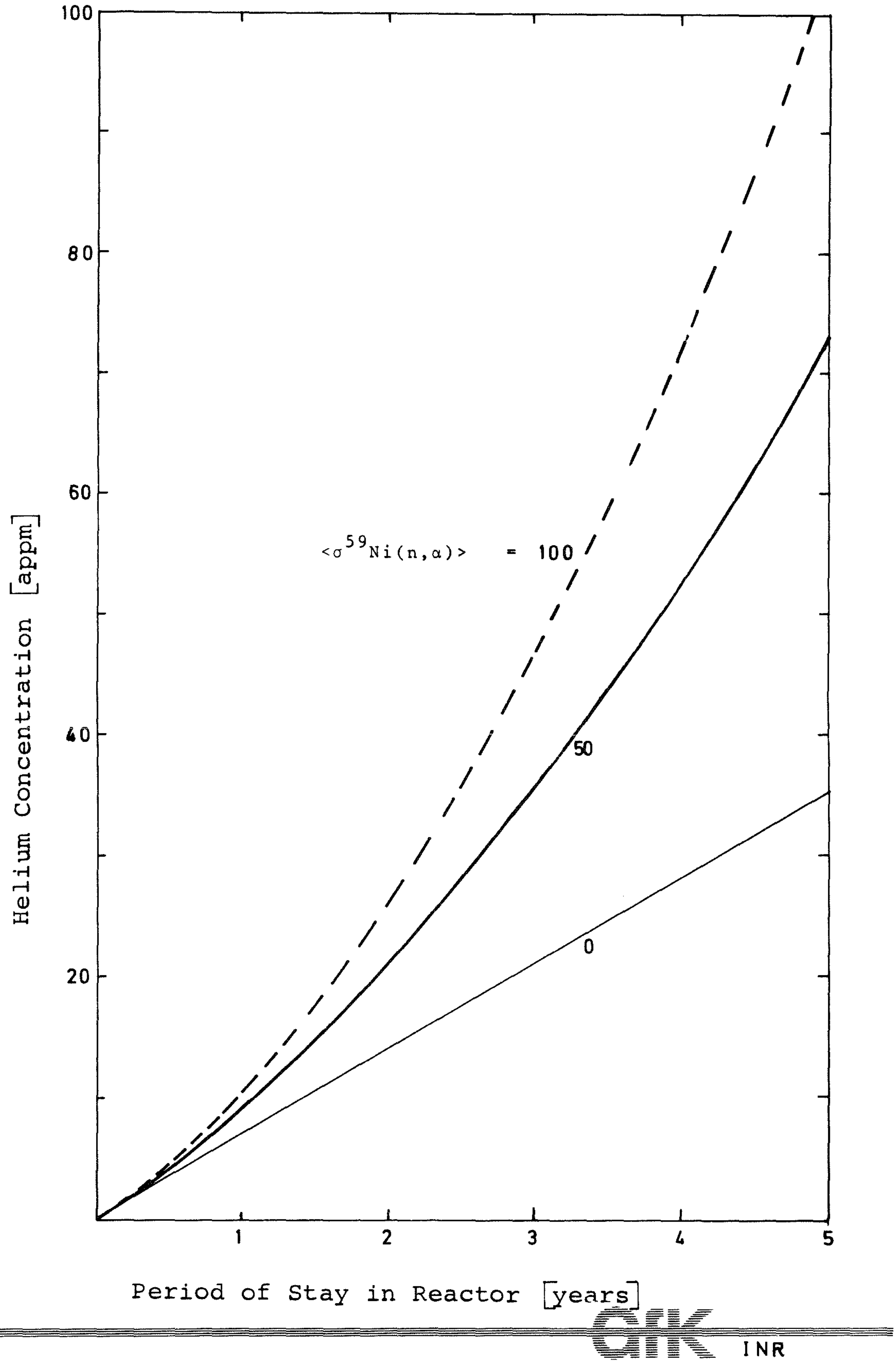


Fig. 7: Sensitivity of the helium generation in stainless steel on the value of $^{59}Ni(n,\alpha)^{56}Fe$ cross section. The lower curve represents the helium production without considering the two step $^{58}Ni(n,\gamma)^{59}Ni(n,\alpha)^{56}Fe$ process. The thick solid line gives the helium production in stainless steel with the cross sections shown in Fig. 5 i.e. $\sigma(n,\gamma)$ = 22,2 mb, $\sigma(n,\alpha)$ = 50.8 mb. The dashed curve with $\sigma(n,\alpha)$ = 100 mb.

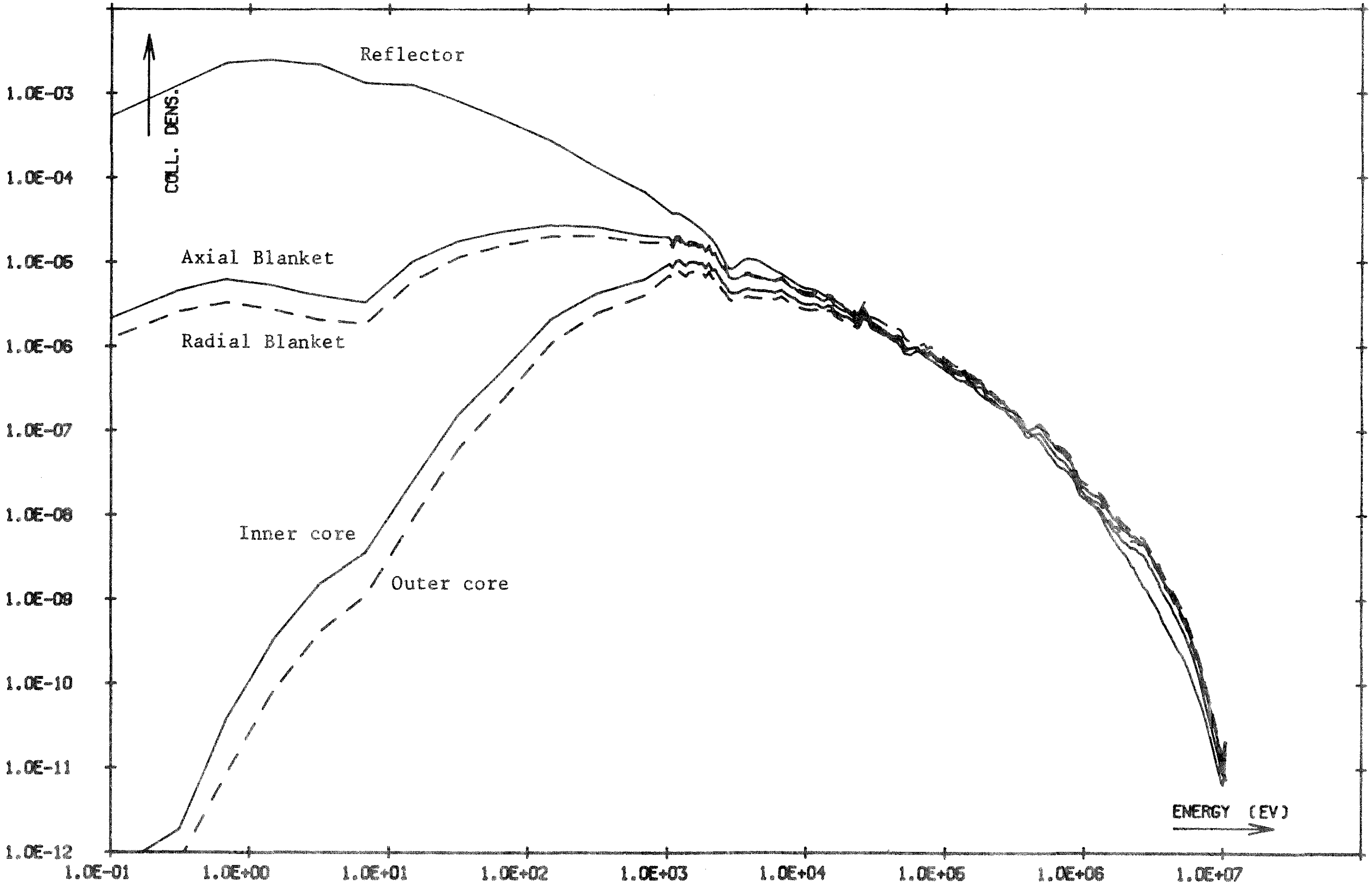


FIG. 8 ZONE COLLISION DENSITIES FOR NEACRP BENCHMARK