A First Approach to Data Needs and Target Accuracies for Hybrid Systems

M. Salvatores
Commissariat à l'Energie Atomique/DRN, Cadarache
(France)

Abstract

Feasibility studies for hybrid systems (i.e. a particle accelerator coupled to a subcritical multiplying medium) need a large amount of data, in a large energy range, whatever the differences in the applications.

However, data needs, uncertainties impact and design parameter target accuracies have not yet been systematically investigated.

The present paper is a first attempt in this field, in particular in relation to intermediate energy (i.e. $E \geq 20$ MeV) data and intranuclear cascade codes.

1 - Introduction

Hybrid systems which couple a particle accelerator (usually a proton accelerator) to a subcritical medium, already studied in the past [1, 2], have been recently investigated with new objectives (see for example Refs. 3, 4 and 5). The physics of these system has been the object of detailed analysis (see for example Ref. 6). In general, a proton beam interaction with a target (e.g. lead target) produces a large amount of neutrons which can be used as such (i.e. as spallation neutron source), or as a neutron source to be injected into a multiplying system with a number of different objectives :

- radioactive waste transmutation (actinides and/or long-lived fission products) [3, 4, 7, 8],
- plutonium burning [3],
- energy production [3, 5].

The target can be separated from the multiplying medium, or the target can be the multiplying medium itself [9].

Most of the feasibility studies have been based on what we will call the "standard" approach :

- Since the proton energies considered in present studies are generally in the range ~ 0,8 ÷ 2 GeV, intranuclear cascade (INC) codes have been used to produce an energy neutron source below ~ 20 MeV, which is successively propagated by standard neutron transport codes (deterministics S_N, or, more often, Montecarlo). Interaction cross-sections for $E \leq 20$ MeV are taken from the existing standard evaluated nuclear data files.

This type of calculation scheme, allows to evaluate the main parameters of interest of hybrid systems, whatever the application :

- The energy balance (e.g. the fraction of energy produced in the subcritical medium that has to be fed back to the accelerator).
- The neutron balance in the subcritical medium (directly related e.g. to the transmutation potential of the system, via the excess neutron production).
- Spallation products and their toxicity.
- Damage and activation of structures.

- Coupling of accelerator beam and target (e.g. energy deposition, neutron backscattering etc ...).
- Shielding.

A major question is related to the predictive power of the different calculation systems which have been used by the different groups and what level of confidence (or, better, what uncertainties) can be associated to the announced performance parameters.

A first step in this direction has been the NEA - Nuclear Science Committee initiative to launch Code intercomparisons in the field of the prediction of intermediate energy data (i.e. the benchmark related to protons interacting with thin targets [10]) and of INC codes (i.e. the benchmark related to protons interacting with thick targets [11]).

This exercise, mainly, but not exclusively, related to the assessment of the performance of different systems for the nuclear waste transmutation, should provide a first indication of the predictive power of the different codes, problem areas to be addressed by code improvements, or new measurements. Moreover, indications are expected on the possible need for an alternative calculation route, i.e. :

- INC codes provide a neutron source for transport calculations below E ~ 100 MeV. New evaluated data would then be necessary between ~ 20 MeV and 100 MeV.

This procedure will avoid the need to upgrade present INC codes in the region of pre-equilibrium and presents the advantage to extend the separation of data from transport codes up to ~ 100 MeV from the present limit of ~ 20 MeV. Codes to evaluate data in that energy range exist and file formats can be defined. A detailed discussion on this problem, together with a first state of the art on data compilations can be found in Refs. 12 and 13.

2 - The definition of uncertainties and target accuracies

The assessment of data uncertainties is the first step in a logical process intended to point out needs for better data and/or new measurements. It has been a standard procedure in establishing the interface between basic nuclear data and fission reactor physics calculations, to fold data uncertainties with the sensitivity coefficients of the main design parameters to assess the uncertainty associated to these parameters due to data uncertainties. As a further step, if design (integral) parameters target accuracies are defined, one can define the level of the required accuracies on data, as it was done e.g. in Refs. 14 and 15, and, more recently, in Ref. 16 ; where a study was performed on required nuclear data accuracies to meet requirements on the radiotoxicity source reduction via waste transmutation in fission reactors.

In this procedure, an essential elements is the definition of sensitivity coefficients for the major parameters of interest I_j, expressed in an integral form. These sensitivity coefficients are in general dependent of the specific configuration being considered.

At present for hybrid systems, sensitivity analysis have not been done in a systematic way.

Some studies have been done by Cierjacks [17] and Atchinson has addressed these issues in relation with a spallation source design [18].

For a systematic approach, one is lacking generalized sensitivity formulations to quantify $(\delta I_j/I_j)/(\delta\sigma_k/\sigma_k)$.

However, we will try to give some preliminary indications in the following paragraphs.

3 - Some data requirements for hybrid systems

In a qualitative sense, one can try to relate a particular class of data and their uncertainties to a particular class of hybrid system parameters.

Examples of such relations are :

- The total neutron yield (number of neutrons/proton) and the system energy balance.
- The high energy component of the distribution of the neutron source ($E \geq 20$ MeV) is related (among other parameters) to structure damage and activation in an energy range not much explored up to now.
- The low energy ($E \leq 20$ MeV) component of the neutron source distribution is related, via the spectrum averaged neutron interaction cross-section $\bar{\sigma}$, to the neutron balance in the subcritical medium.
- The neutron angular distributions are related to the assessment of the neutron backscattering and to the shielding performance.
- The spallation product distributions in (A,Z) are related to the radiotoxicity balance of the system and of its fuel cycle.

Starting from these (and more) qualitative statements, we have tried to extract some preliminary quantitative indications for a few relevant parameters.

3.1 - The energy balance

In Reference 6, it was indicated a simple relation between the fraction f of the energy produced in the subcritical region of an hybrid system which is fed back to the accelerator, the subcriticality level (1/Keff - 1), and the number Γ of neutrons (per one fission), returning to the subcritical region via the spallation process :

$$f = \left(\frac{1}{K_{eff}} - 1 \right) \frac{\nu}{\Gamma} \qquad (1)$$

where ν is the number of prompt neutrons emitted per fission

Γ is directly proportional to the neutron yield per proton, Z. Then it is clear that :

$$\delta f / f = - \frac{\delta Z}{Z}$$

Also, if i is the proton beam current, one has [6] :

$$\delta i/i = -\frac{\delta Z}{Z}$$

From this simple direct proportionality, it is possible to indicate a requirement on Z (on or the nuclear mechanisms which define the neutron yield) of an accuracy of the order of 10 ÷ 20 %.

On the contrary, the energy balance (e.g. the f parameter) seems to be very little affected by the source neutron spectrum in energy. As an example, if one propagates a proton beam (E_p = 1.6 GeV) into a subcritical medium (e.g. a molten salts fuel) at different levels of subcriticality, and one modifies artificially the spectrum of the neutrons which are propagated below 20 MeV keeping constant the number of neutrons produced per proton, the effect on f is fairly small, as indicated in the following table :

n Source spectrum (below 20 MeV)	f parameter (Ref. 6)	
	$K_{eff} = 0.85$	$K_{eff} = 0.95$
1 - Evaporation spectrum from HETC (Reference)	0.57	0.18
2 - As above, but fraction at E > 6 MeV multiplied arbitrarily x 2	0.56	0.18
3 - Fission spectrum replacing evaporation spectrum	0.59	0.19

As an example, the difference between the fission spectrum and the evaporation spectrum for the PHOENIX system [9] as calculated by KADI et al [19], is given in figure 1.

3.2 - The neutron balance in the subcritical region

In a subcritical system, with external source S_{ext}, a neutron surplus is available (e.g. to be used for transmutation) if :

$$G = S_{ext} - D_{fuel} - (L + CM) > 0$$

D_{fuel} : nuclear fuel neutron "consumption"/fission (a negative value means "production")
L : neutron leakage/fission
CM : neutrons absorbed in parasitic captures/fission

For any isotope J, D_J is function of average cross-sections ($\overline{\sigma}_f, \overline{\sigma}_c, \overline{\sigma}_{n,2n}$ etc), and decay constants :

$$D_J = \sum_{J1_i} P_{J \rightarrow J1_i} \left\{ R_{J1_i} + \sum_{J2_k} P_{J1_i \rightarrow J2_k} \times \left[R_{J2_k} + \sum_{J3_n} P_{J2_k \rightarrow J3_n} (\ldots) \right] \right\} \quad (2)$$

where $P_{JN_r \rightarrow (JN+1)_s}$ = probability of the JN_r nucleus transmuting into a $(JN+1)_s$ nucleus ; that is, the ratio of the rate of direct conversion $JN_r \rightarrow (JN+1)_s$ and the total rate of all the possible processes for JN_r

R_x = neutron loss as a result of the appearance of x from the previous generation.

For example,

$$R_x = \begin{cases} 1 & \text{for a transmutation by neutron capture reaction} \quad , \\ 0 & \text{for decay}, \\ 1-\nu & \text{for fission} , \\ -1 & \text{for (n,2n) reaction} . \end{cases}$$

The sensitivity of the neutron balance (and of G) to the neutron source spectrum is relatively small (both in the case of a target separated from the multiplying subcritical region, or in the case that the target is that region itself). In fact, the neutron spectrum in the subcritical region is dominated by the effect of the neutron interactions in the region itself, and it will become very close to the spectrum typical of that region in absence of external source.

The sensitivity of the D_J parameters to the neutron spectrum is directly related to the sensitivity of the average cross-sections $\overline{\sigma}$ to the weighting spectrum, and this is relatively small, except for some (n,2n) or threshold fission cross-sections. An example is given in Ref. 19. The G parameter, which integrates all these effects will only be marginally affected by the "external" neutron source spectrum.

The neutron balance will be mostly affected by the uncertainties of neutron cross-sections below 20 MeV, i.e. data which have already been tested in most standard fission reactor applications.

3.3 - Neutron damage

It has been argued (see for example ref. 17), that the presence of a high energy ($E_n \geq$ 20 MeV) component in the neutron spectrum likely to be issued by spallation in a thick target (a fraction of up to 10 %) can produce a potential and very significant effect of damage of the structures which will be irradiated by such neutrons.

It appears then that a good knowledge of this high energy component will be highly desirable.

In the past, similar problems were studied in the context of fusion reactor dosimetry (see for example Ref. 20).

To get a feeling of the impact of the uncertainties on the high energy component, one can make use of the notion of "displacement" cross-section σ_{dpa}, defined as [21] :

$$\sigma_{dpa} = \sum_i \sigma_i (E) \int_{E_{p,min}}^{E_{p,max}} \nu(E_p) P_{E_i} (E, E_p) \, dE_p$$

where : σ_i : X-section for neutron interaction of type "i"
E_p : primary energy
P_{E_i} (E,Ep) : probability of obtaining E_p from reaction i
$\nu(E_p)$: number of displaced atoms by a primary of energy E_p
The number of displacements per atom DPA are defined as :

$$DPA = \int_0^{\infty} \sigma_{dpa}(E)\, \phi_n(E)\, dE$$

where $\phi_n(E)$ is the energy spectrum of the neutron flux.

The quantity "DPA" is not used "per se", but it is used as a quantity able to characterise a specific neutron irradiation field. Damage in materials (e.g. swelling in terms of volume V modifications $\Delta V/V$) are the expressed as a function of DPA, but also accounting for other specific characteristics, such as He production due to neutron irradiation, which can modify substantially the dependence of, say, $\Delta V/V$ from DPA. A schematic illustration is given in fig. 2, taken from reference 22. It is clear that the knowledge of the high energy component of the neutron spectrum will enable to characterise an irradiation and to deduce from (mostly experimental) correlations, the potential damage to materials.

In an attempt to be more specific, there is interest to limit the uncertainty on the DPA abscissa of graphs like the one given in fig. 2. In the case of a fission neutron spectrum (as given in fig. 1) and of the σ_{dpa} of Fe, extrapolated above 20 MeV, the average $\overline{\sigma}_{dpa}$ (Fe) is $\simeq 400$ b. In the case of an evaporation spectrum $\overline{\sigma}_{dpa}$ (Fe) $\simeq 700$ b, and the fraction of DPA due to neutrons with $E_n \geq 20$ MeV is approximately 15 ÷ 20 %.

An uncertainty of the order of 30 ÷ 50 % on the high energy ($E_n \geq 20$ MeV) component seems acceptable :

- to define the DPA scale,
- to evaluate the (n,α) reaction rate in the structural materials,
- to evaluate the activation of these materials.

More precise requirements can only come from more detailed and specific feasibility studies.

3.4 - Spallation products toxicity

Spallation target activity evaluation and radiotoxicity balance in a hybrid system, require an evaluation of spallation product distributions in (A,Z).

An example of uncertainties in such distribution, due to potential INC code deficiencies has been illustrated by work at PSI [19]. Switching off the fission model in the HETC code produces a dramatic variation of the spallation product distribution and a strong variation of the associated radiotoxicity (approximately a factor 10).

A factor 2 as target accuracy on the spallation product radiotoxicity and its evolution with time seems to be acceptable, with respect to the total radiotoxicity due to the fuel irradiation.

It is not clear at present to what extent uncertainties an (A,Z) distributions allow to meet this target accuracy.

A first step should be to give preliminary estimates on the (A,Z) distribution uncertainties, may be using simple formulations like the one proposed by Rudstam and given by Atchinson (Ref. 24) :

$$\sigma\,(Z_p\,A_p) = f(A_t,E)\,\exp P(A_t - A_p)\,\exp\,(- R\,|Z_p - Z_{max}|^{3/2})$$

and looking to $\delta\sigma/\sigma$ variations due to parameters P, R etc variations.

4 - Conclusions and preliminary recommendations

A - Target accuracies

1 - Today, few target accuracies can be formulated for hybrid system parameters on sound basis. However, for most feasibility studies, a $\pm 10 \div 20$ % uncertainty on the energy balance (in the sense indicated in the present paper) can be tentatively indicated. This target uncertainty would require a corresponding $\pm 10 \div 20$ % accuracy on the total neutron yield, as given e.g. by standard INC codes when dealing with proton spallation in thick targets.

2 - A target accuracy of ± 20 % on parameters related to neutron damage (e.g. α-production by neutrons, DPA parameter for irradiation characterisation etc) can require an accuracy of approximately $\pm 30 \div 50$ % on the high energy ($E_n \geq 20$ MeV) component of the neutron spectrum, in the hypothesis that it represents approximately 10 % of the total evaporation spectrum.
Complementary requirements in this field are obviously (n,α) cross-sections at $E_n \geq$ 20 MeV for most structural materials and an appropriate modelling of σ_{dpa} at $E_n \geq$ 20 MeV [17, 20].

3 - For the spallation product radiotoxicity source term and its evolution with time, at the present stage of studies, a ± 100 % target accuracy can be tentatively indicated.
Much work seems to be necessary to establish the predictive power of codes in this field and experimental validation seems to be widely needed.

4 - Shielding studies associated to hybrid system, will play a relevant role. However, traditionally target accuracies for doses around the installations are related to regulation requirements and conservative uncertainty evaluations are associated to relevant calculations. It is then required to express uncertainties even if tentative values are used (e.g. in neutron yield angular distributions) in order to evaluate potential uncertainties on shielding parameters.

5 - A possible target accuracy of ± 10 % can be indicated on the neutron balance in the subcritical medium (and then e.g. on the G neutron "excess" parameter defined in Ref. 6). The uncertainty on the neutron "excess" per fission is directly related to the transmutation potential of an hybrid system (Ref. 7). Since the neutron balance uncertainty is mostly related to standard neutron interaction data below 20 MeV, new requirements will not be expressed except for specific materials not yet used in fission reactors or resulting from spallation, and for which data are not generally available in current evaluated data libraries. These needs have to be specified in the frame of each specific project [24] and do not concern in principle INC codes or intermediate energy data.

B - Present methods : predictive power and evolution

The predictive power of some of the present methods and codes has been roughly indicated [18, 25, 26].

More work is needed to have rough estimates on :

- neutron yields,
- proton, α etc yields,
- high energy (20 MeV $\leq E_n \leq$ 100 MeV) neutron spectrum (in angle and energy).

If explicit statements cannot be made, it would be relevant to establish simple relations with, say, double differential cross-sections uncertainties.

- spallation product distribution in (A,Z).

These estimates will be of interest for target material candidates (Pb, W, but also U or Th).

With respect to the present standard calculational approach (i.e. INC codes producing a neutron source to be propagated below 20 MeV), improved treatment in INC codes of the physics in the region 20 ÷ 200 MeV should be the first priority, since this approach will probably stay the preferred approach for most feasibility studies.

However, possible future optimization or more detailed studies could require the evolution towards a scheme in which INC codes are used to produce a neutron source to be propagated below, say 100 MeV.

In that case evaluations should be needed from 20 MeV to ~ 100 MeV [13]. A pilot project should be envisaged, mostly focused on a structural material (Fe) and a fissile material (U or Th), to produce a evaluated files to be used in appropriate versions of transport codes (S_N or MonteCarlo), which will also require adaptation (in particular if particles other than neutrons and photons have to be handled).

C - Experimental validation

The present meeting should hopefully express needs for further validation work, in particular needs for new experiments.

It seems, from the viewpoint of the hybrid system feasibility studies, that the highest priority is related to spallation product distribution measurements with acceptable experimental uncertainties.

A few fundamental, high precision, measurements of double differential cross-sections, with proton energies between 0.5 and 2 ÷ 3 GeV, could benefit the validation of INC codes and codes which can potentially be used to evaluate data between 20 and 100 MeV.

References

1 - W.B. Lewis, Report AECL-968, Chalk River, Ontario, 1952

2 - H. Takahashi and H. Rief. "Concepts of Accelerator - Based Transmutation Systems". Proc. Specialists' Meeting Accelerator - Based Transmutation, Villingen, Switzerland March 24-26, 1992.

3 - C.D. Bowman et al. "Nuclear Energy Generation and Waste Transmutation Using an Accelerator. Driven Intense Thermal Neutron Source". LA-UR-91-2601 (1992).

4 - T. Takizuka "Overview on Nuclear Design Problems of Accelerator - Based Transmutation systems with Emphasis on Target Facilities and their Interface with Accelerators". Proc. Specialists' Meeting, Villingen, March 24-26, 1992.

5 - F. Carminati et al., Report CERN AT/93/-47/ET (1993).

6 - M. Salvatores, I. Slessarev, M. Uematsu, Nucl. Sci. Eng. 116, 1 (1994).

7 - M. Salvatores et al. "Nuclear Waste Transmutation : Physics Issues and Potential in Neutron Fileds". Int. Reactor Physics Conf., Tel-Aviv, January 23-26, 1994.

8 - W.C. Saller et al. "Comparison of accelerator-based with reactor-based waste transmutation schemes" ibidem.

9 - G.J. van Tuyle et al. "The Phoenix Concept". BNL-52279 (1991).

10 - P. Nagel and M. Blann "Specifications for an International Code and Model Intercomparison for Intermediate Energy Reactions" NEA/NSC/DOC 3 (1992).

11 - D. Filges and H. Küsters "Benchmark specifications for Intercomparison of Thick Target Calculations for Transmutation Purposes" NEA/NSC/DOC 14 (1992).

12 - A.J. Koning, NEA/NSC/DOC (92) 12.

13 - A.J. Koning, NEA/NSC/DOC (93) 6.

14 - L.N. Usachev et al. "Determination of Transactinide Data Required Accuracy for Burn-up Calculation in Fast Reactors". Proc. Int. Conf. on Neutron Physics and Nucl. Data, Harwell (1978).

15 - M. Salvatores and G. Palmiotti, Nucl. Sci. Eng. 87, 333 (1984).

16 - G. Palmiotti, M. Salvatores and R. Hill, Nucl. Sci. Eng., August 1994.

17 - S. Cierjacks "Cross Section Measurements, Integral Experiments Validation of Importance to Accelerator - Based Transmutation. Proc. Specialists' Meeting on Accelerator - Based Transmutation, Villigen, Switzerland 24-26 March 1992.

18 - F. Atchinson "Data and Methods for the design of Accelerator - Based Transmutation systems". ibidem.

19 - Y. Kadi, P. Wydler "Neutronic Analyses of Hybrid systems with TRU targets". This meeting.

20 - See e.g. "Nuclear Data for Radiation Damage Estimates" INDC (NDS) - 179G (1986).

21 - M.J. Morgett, M.T. Robinson, I.M. Torrens, Nucl. Eng. Design, 33, 50 (1975).

22 - D. Smith "Neutron Dosimetry for Radiation Damage in Fission and Fusion Reactors". Proc. Conf. Nucl. Cross-sections for Technology, Knoxville (1979).

23 - F. Atchinson. "Aspects of Calculational Needs for Spallation Facilities". This meeting.

24 - E.D. Arthur. "Nuclear Data Needs for Accelerator - Driven Transmutation Systems" ibidem.

25 - M. Blann. "Evaluation of LLNL-ALICE Code Contribution". This meeting.

26 - P.G. Young and M.B. Chadwick. "Calculation of Proton and Neutron Emission Spectra with the GNASH Code". This meeting.

Figure 1. **Comparison between evaporation spectrum and fission spectrum in the PHOENIX target model (oxide fuel)**

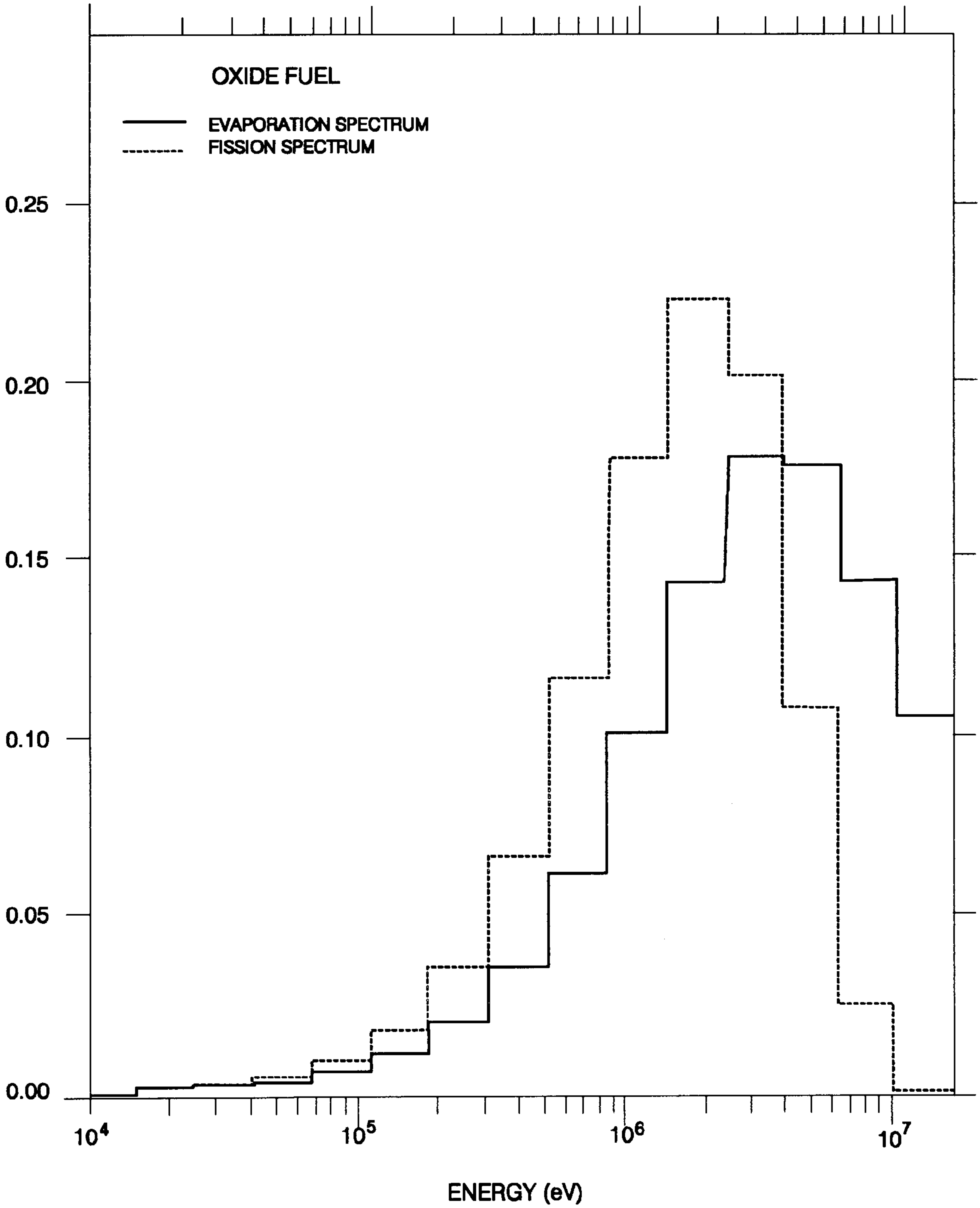
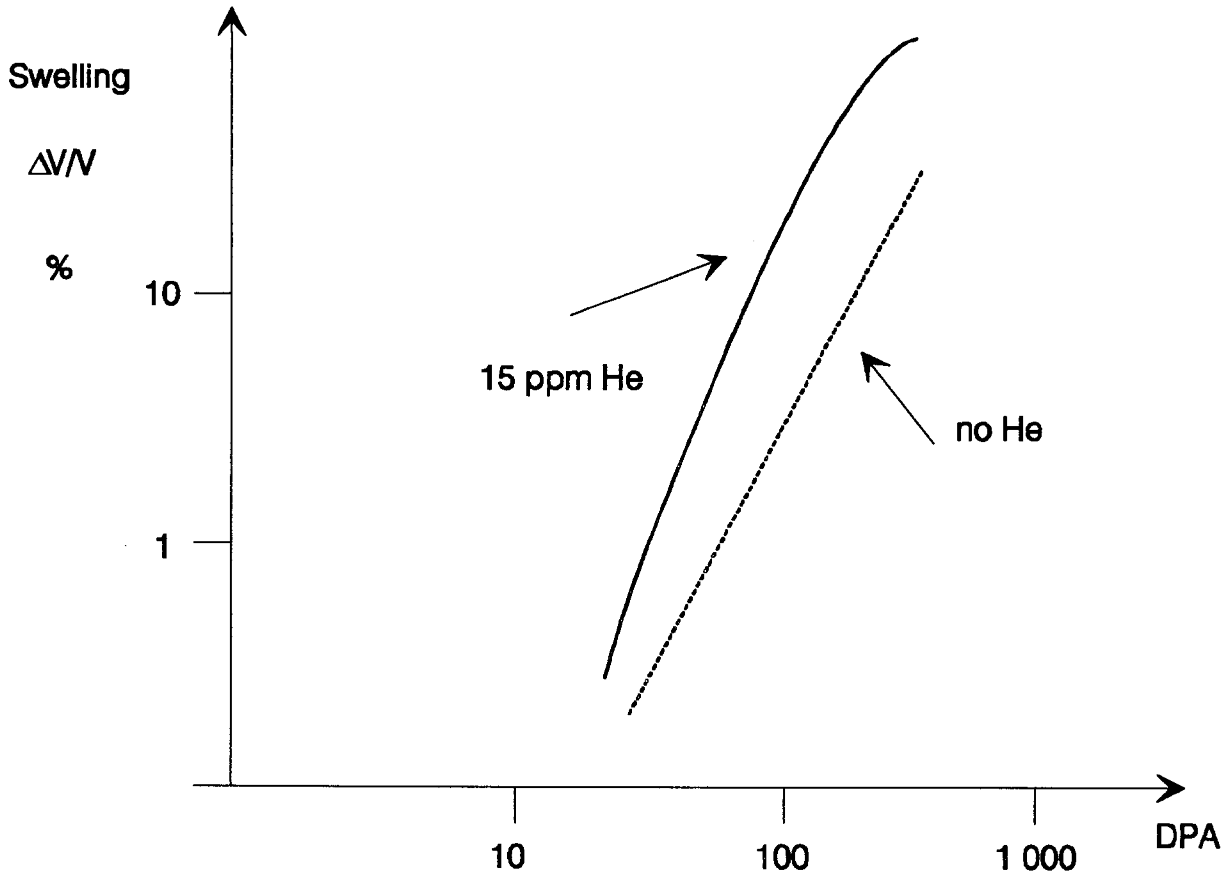


Figure 2. **Relation between swelling and DPA (stainless steel)**


Swelling
ΔV/V
15 ppm He
no He
DPA