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Radiation Damage Calculations for Compound Materials

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Abstract

Displacement damage calculations can be performed for 40 elements in the energy range up to 20 MeV with the SPECTER computer code. A recent addition to the code, called SPECOMP, can intermix atomic recoil energy distributions for any four elements to calculate the proper displacement damage for compound materials. The calculations take advantage of the atomic recoil data in the SPECTER libraries, which were determined by the DISCS computer code, using evaluated neutron cross section and angular distribution data in ENDF/B-V. Resultant damage cross sections for any compound can be added to the SPECTER libraries for the routine calculation of displacements in any given neutron field. Users do not require access to neutron cross section files. Results are presented for a variety of fusion materials and a new ceramic superconductor material. Future plans and nuclear data needs are discussed.

Introduction

Fundamental radiation damage calculations involve the determination of atomic recoil distributions induced by neutron irradiations. These primary knock-out (pka) energy distributions depend on the neutron cross sections and angular distributions as a function of neutron energy. We perform such calculations using the DISCS[1] computer code and using evaluated neutron data from the ENDF/B-V file.[2] DISCS results for yttrium are shown in figure 1, where the relative contributions from each reaction channel are displayed. The further calculation of total atomic displacements requires a model for the partition of recoil atom energy into nuclear and electronic energy loss events and a model for the production of secondary displacements caused by nuclear collisions and reactions. We have assumed the Lindhard[3] model, with modifications suggested by Robinson.[4] The DISCS code calculates the recoil atom energy distributions for each nuclear reaction given in ENDF. Angular distribution data is always given explicitly for elastic scattering and an evaporation model is assumed for other reactions, except for the (n,xn) reactions where a Monte Carlo calculation is needed to determine the proper recoil distributions. For charged particle emission, the evaporation model is centered on the coulomb barrier. Displacements are also included for (n,γ) reactions and for beta decay. It should be noted that our calculations of the displacements per atom (dpa) only describe the fundamental nuclear interactions and hence are useful as an exposure index which is independent of the neutron spectrum. However, dpa may not be necessarily related to materials property changes since many of the displacements may recombine with holes in the lattice and additional molecular dynamic calculations are needed to determine stable defects.

The calculated displacement damage cross sections and recoil energy atom distributions are stored in master libraries. The SPECTER computer code[5] is then used to calculate displacements and recoil distributions for any given neutron spectra. SPECTER also contains libraries for gas production[6] and

total energy deposition. Hence, users will obtain a comprehensive set of damage parameters for 40 different elements for any specified neutron irradiation and they only need to specify the neutron spectra and length of irradiation.

SPECOMP Calculations

The calculations from SPECTER only consider pure elements. In order to calculate displacements from compound materials, the procedures must be modified to include all combinations of recoiling atoms and matrix atoms. For example, if we consider Li_2O, then we must calculate the probabilities that Li atoms will displace O atoms as well as Li atoms and that O atoms will displace both Li and O atoms. The relative contribution from each of these combinations is shown in figure 2. These probabilities are determined by the Lindhard equations[3] which are usually only seen in the reduced self-ion form. These effects have been discussed by Parkin and Coulter.[7]

When considering damage in compounds, it is important to consider the possible atomic distribution of elements. In a true chemical compound, the probability that a recoiling atom will interact with a given species simply depends on the atomic abundance and this is the approximation which we have assumed in SPECOMP. However, if an alloy contains clumps or segregation of elements, then the present calculations are not necessarily appropriate and it might be better to sum the damage for the individual elements or to look at local damage by region in the material.

Secondary displacements are taken to be zero if the recoil energy is below a threshold energy, one from threshold up to twice that energy, and equal to the damage energy divided by twice the threshold at higher energies. For self-ion damage, appropriate threshold energies are recommended by the ASTM.[8] Unfortunately, there is no such agreement as to suitable values for the threshold energies for compound materials. This presents us with a serious problem since SPECOMP calculations are found to be very sensitive to our choice of threshold energies. When considering self-ion damage, the threshold can be changed after the calculations by simply renormalizing the results. This is not the case in compound calculations and it becomes meaningless to report damage energy cross sections in units such as keV-barns. Thus, a new calculation must be performed for each choice of threshold energy. The threshold for a given element will also differ according to the compound since the value really depends on the interatomic potentials for a given chemical species.

SPECOMP calculations can be performed rather quickly since they take advantage of the recoil atom energy distributions stored in the SPECTER libraries. These pka distributions only depend on the interaction of neutrons with a given element and hence do not have to be recalculated for different compounds. The pka data are stored in a 100 group energy structure at each of 100 incident neutron energies. Hence, there are 10,000 entries for each element. The SPECOMP code integrates over the allowable range of recoil energies at each of the 100 neutron energies for each combination of recoil and matrix atom. The results are then normalized by atomic abundances in each compound and summed to produce a singe displacement cross section value at each neutron energy.

Results

Displacement cross sections calculated with SPECOMP are usually added to a library in SPECTER for routine use, as described previously. SPECOMP also lists secondary displacement calculations and separate displacement cross sections for each element in the compound. This may be important to the user since the net dpa cross section conveys no information regarding the type of atoms which are being displaced. SPECOMP does not need to list pka recoil energy spectra since this information is already available for each element in SPECTER.

SPECOMP calculations have been completed for fusion breeder materials, such as Li_2O and $LiAlO_2$; insulators, such as Al_2O_3, SiO_2, and TaO; engineering alloys, such as 316 stainless steel, NbTi, and V-15Cr-5Ti; research materials, such as Cu_3Au; and the new ceramic superconductor $Y_1Ba_2Cu_3O_7$. In lieu of accurate knowledge of appropriate threshold values, we simply used the same thresholds for the compounds as we normally use for the pure elements. This has the advantage of facilitating comparisons with previous approximations of damage in compounds which simply sum elemental dpa values weighted by atomic abundances. Calculations for several compounds are listed in Table I and compared with elemental calculations in figures 3-5.

As can be seen in Table I and figures 3-5, there may be sizable differences (eg - LiO_2, $LiAlO_2$, and TaO) between the SPECOMP calculations and damage calculated from a weighted elemental sum. This is shown more clearly in figure 6 which displays the ratio of the damage calculated with SPECOMP divided by a weighted sum of elemental damage. In some cases, such as stainless steel and the vanadium alloy, the results of both calculations are nearly identical. Three factors appear to be important in determining whether or not SPECOMP results differ substantially from elemental sums, namely, differences in atomic mass, differences in threshold energies, and differences in cross sections. These effects can be seen in figure 2 which shows that more displacements result when Li is the secondary atom rather than O; O displaces more Li atoms than when Li displaces O atoms; and the effect of the 250 keV resonance in the ^6Li cross section is clearly seen. As a general rule, SPECOMP calculations show increasingly larger effects as differences in mass, threshold energy, and cross sections increase between the elements in the compound.

Future Work

SPECOMP is available to SPECTER users and can be easily added to the computer code package. Calculations can be run for any combinations of four elements from the 40 elements now contained in SPECTER. Compounds with more than four elements would require expansion of the matrices in SPECOMP. Calculations with other elements require DISCS calculations and additions to the SPECTER libraries prior to running SPECOMP.

Calculations are now being run for a variety of compounds and recommended dpa cross sections will be added to SPECTER. In this case, users need not run SPECOMP directly unless they wish to change the threshold energies. Of course, the largest uncertainties in such calculations remain the assignment of appropriate threshold energies and further work is needed in this area.

Evaluated nuclear data is now becoming available for ENDF/B-VI. Data formats will be different from ENDF/B-V and recoil atom energy distributions may be given explicitly. Consequently, existing damage codes must be revised to accept the new data prior to recalculating with version VI. There is also a need to extend the calculations to additional elements and to higher neutron energies, especially for accelerator-based neutron sources.

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Figure Captions

1. Illustration of DISCS displacement damage calculation for yttrium where the contribution from each major reaction is shown separately.

2. Displacement damage cross sections from SPECOMP are shown for Li_2O. Each combination of recoiling atom and matrix atom is shown separately.

3. Comparison of dpa cross sections for $LiAlO_2$ calculated by SPECOMP (solid line) with a weighted sum of elemental damage (dotted line).

4. Calculated damage cross sections for 316 stainless steel. In this case there is no difference with the weighted sum of elemental damage.

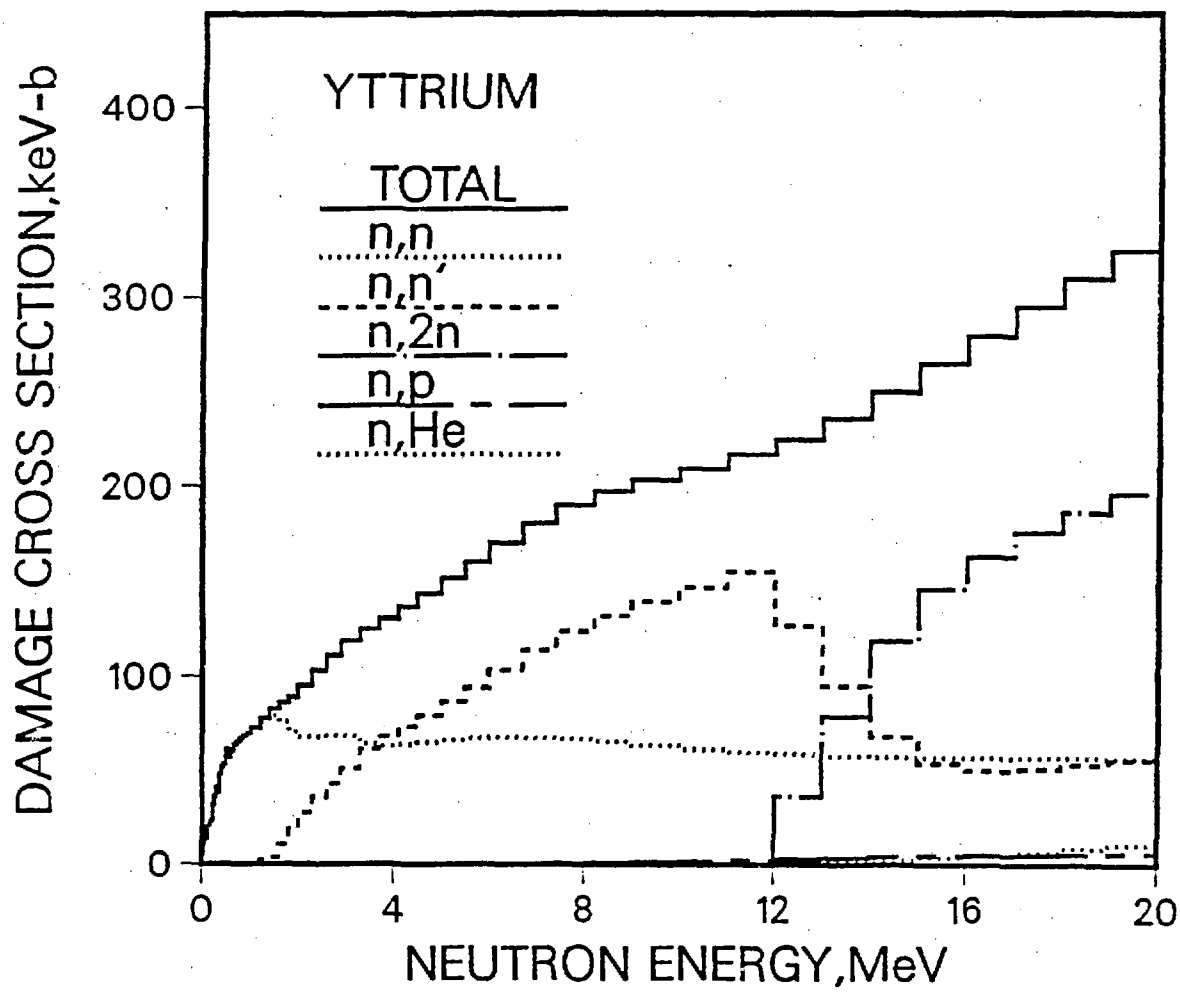
5. Calculated damage cross sections for the new ceramic superconductor Y1-Ba2-Cu3-O7. SPECOMP results (solid line) are compared with the weighted sum of elemental damage (dotted line).

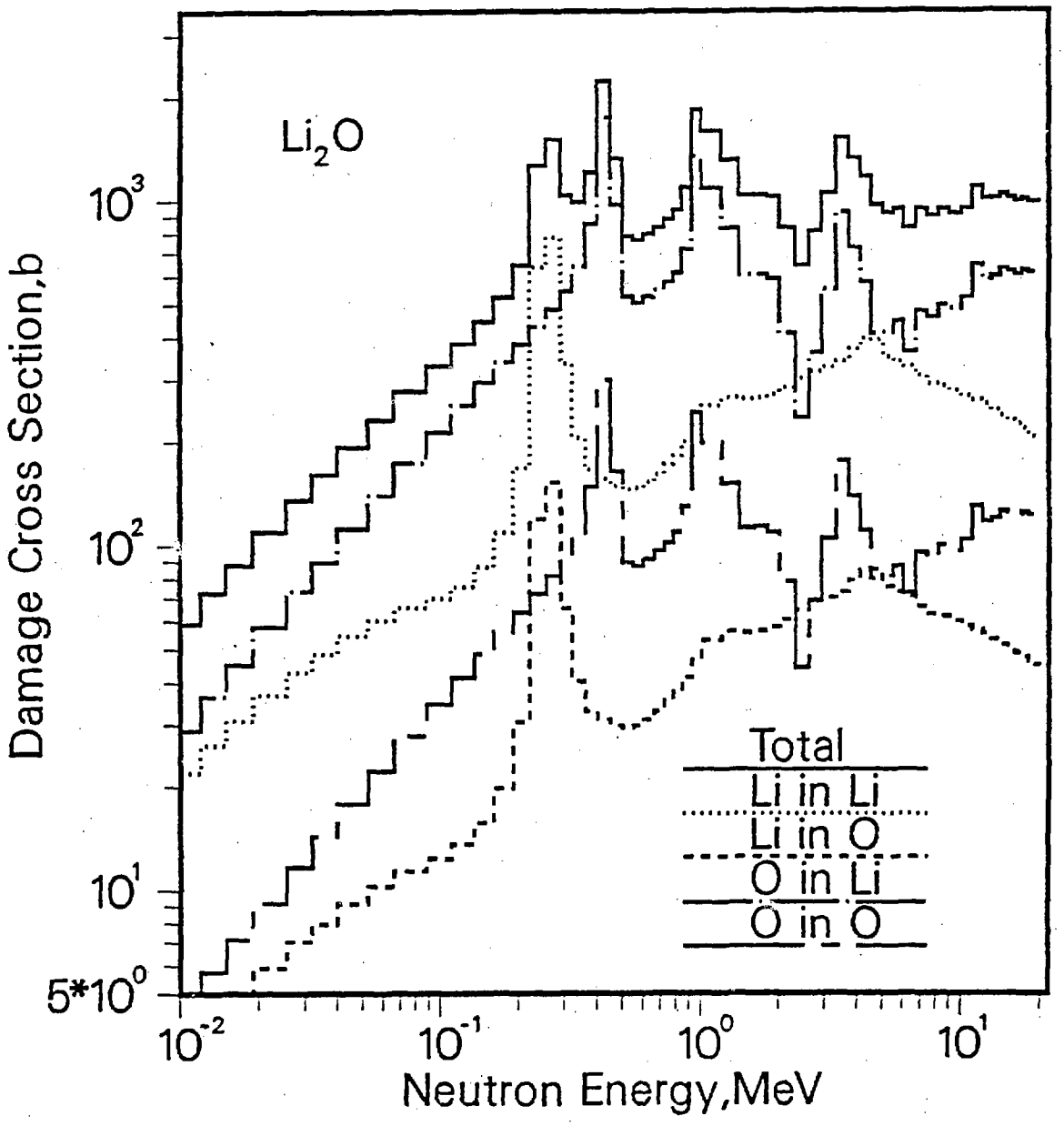
6. Ratios of damage calculated with SPECOMP divided by the weighted sum of elemental damage are shown for various compounds.

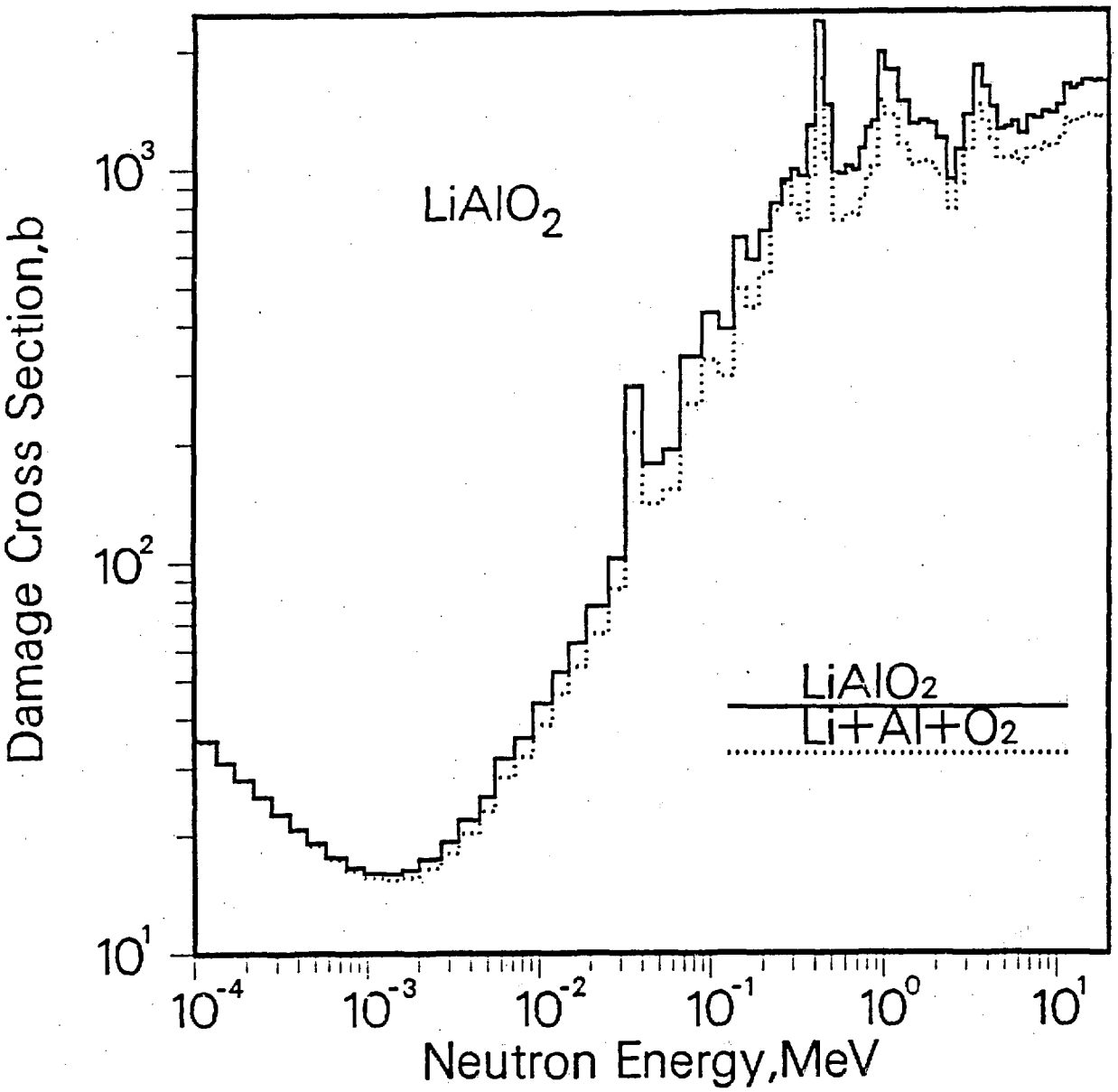
Table I: DPA Ratio (SPECOMP/SPECTER)

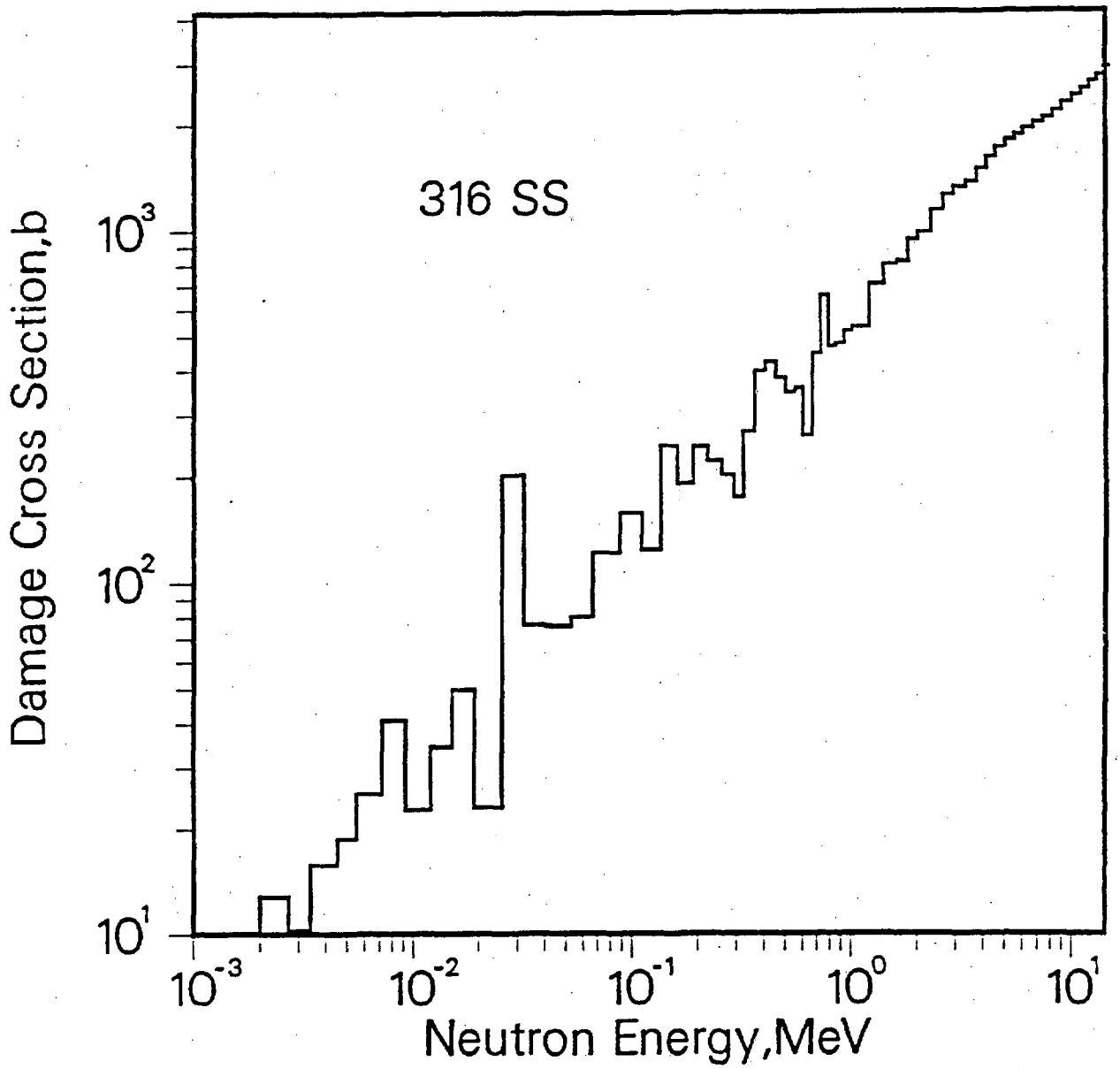
Compound	Fission[a]	14 MeV	STARFIRE[b]	HFIR[c]	FFTF[d]
Li_2O	1.43	1.43	1.31	1.04	1.47
$LiAlO_2$	1.26	1.23	1.25	1.07	1.28
Al_2O_3	0.99	0.98	0.99	1.00	1.00
SiO_2	0.99	0.96	0.99	1.00	1.00
V-15Cr-5Ti	1.00	1.00	1.00	1.00	1.00
316 SS	1.00	1.00	1.00	1.00	1.00
Cu_3Au	0.98	0.98	0.98	0.99	0.99
Nb-Ti	0.99	1.00	0.99	0.99	0.99
TaO	0.89	1.16	0.96	0.86	0.80

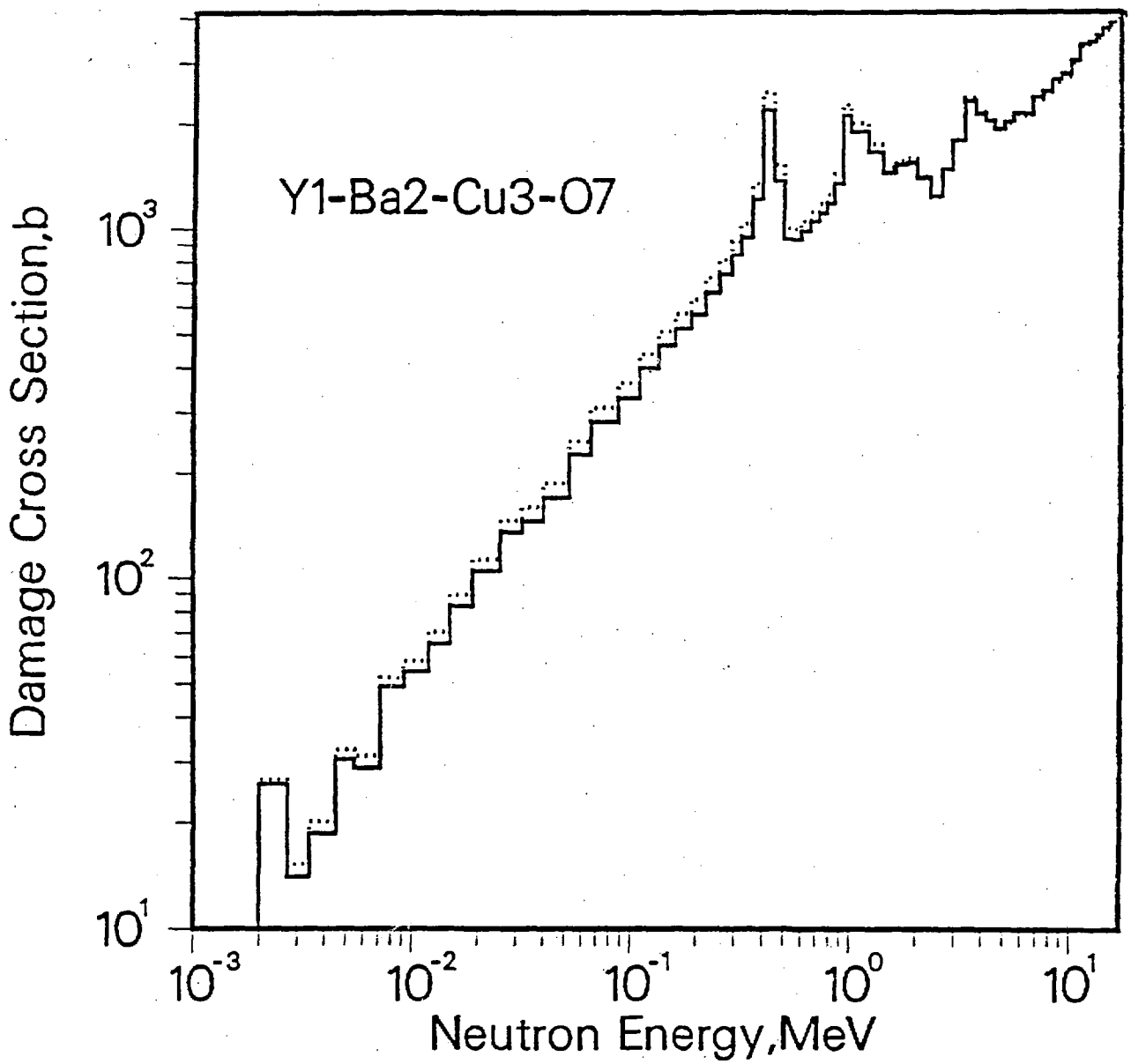
[a] ^{235}U fission spectrum
[b] STARFIRE, first wall spectrum (Ref. 9)
[c] High Flux Isotopes Reactor, PTP, midplane (Ref. 10)
[d] Fast Flux Test Facility, mota, midplane (Ref. 11)

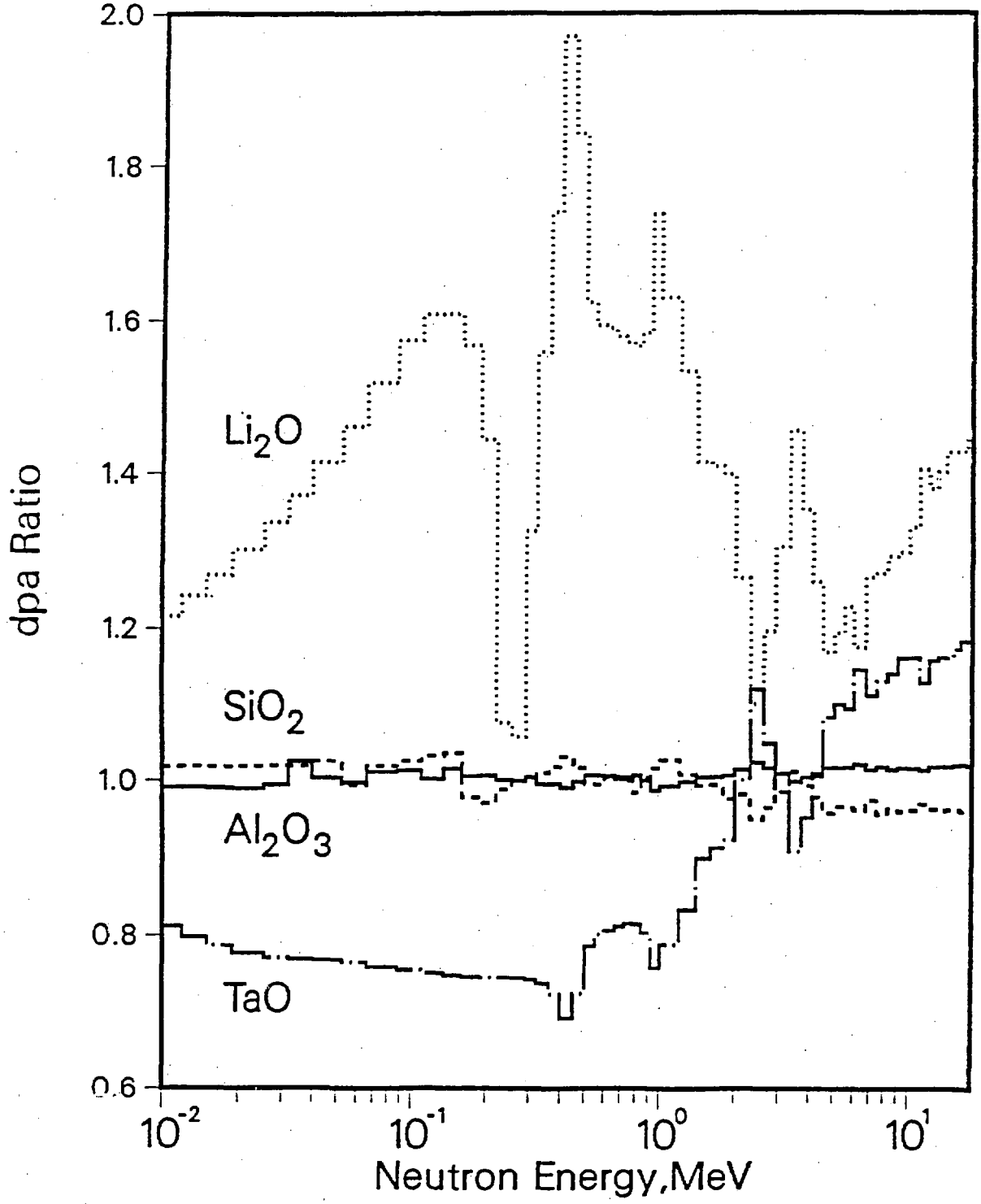

YTTRIUM
TOTAL
n,n
n,n'
n,2n
n,p
n,He
DAMAGE CROSS SECTION,keV-b
400
300
200
100
0
0
4
8
12
16
20
NEUTRON ENERGY,MeV


Li2O
Damage Cross Section,b
10^3
10^2
10^1
5*10^0
10^-2
10^-1
10^0
10^1
Neutron Energy,MeV
Total
Li in Li
Li in O
O in Li
O in O


Damage Cross Section,b
10^3
10^2
10^1
$LiAlO_2$
$LiAlO_2$
$Li+Al+O_2$
10^{-4}
10^{-3}
10^{-2}
10^{-1}
10^0
10^1
Neutron Energy,MeV


316 SS
Damage Cross Section,b
10^3
10^2
10^1
10^{-3}
10^{-2}
10^{-1}
10^0
10^1
Neutron Energy,MeV


Y1-Ba2-Cu3-O7
Damage Cross Section,b
10^3
10^2
10^1
10^{-3}
10^{-2}
10^{-1}
10^0
10^1
Neutron Energy,MeV


2.0
1.8
1.6
1.4
1.2
1.0
0.8
0.6
dpa Ratio
Li_2O
SiO_2
Al_2O_3
TaO
10^{-2}
10^{-1}
10^{0}
10^{1}
Neutron Energy,MeV