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NEUTRON IRRADIATION FACILITIES FOR FISSION AND FUSION REACTOR MATERIALS STUDIES*

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ABSTRACT

The successful development of energy-conversion machines based upon nuclear fission or fusion reactors is critically dependent upon the behavior of the engineering materials used to construct the full containment and primary heat extraction systems. The development of radiation damage-resistant materials requires irradiation testing facilities which reproduce, as closely as possible, the thermal and neutronic environment expected in a power-producing reactor. The Oak Ridge National Laboratory (ORNL) reference core design for the Center for Neutron Research (CNR) reactor provides for instrumented facilities in regions of both hard and mixed neutron spectra, with substantially higher fluxes than are currently available. The benefits of these new facilities to the development of radiation damage resistant materials are discussed in terms of the major U.S. fission and fusion reactor programs.

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INTRODUCTION

The successful development of energy-conversion machines based upon nuclear fission or fusion reactors is critically dependent upon the behavior of the engineering materials used to construct the fuel containment and primary heat extraction systems. Such materials must maintain their structural integrity and dimensional stability in an environment involving high temperatures and heat fluxes, corrosive media (liquid metals, helium, water, etc.), high stresses (sometimes cyclic), and intense fluxes of neutrons. In both fusion and fission systems, a large fraction of the neutron flux is sufficiently energetic to displace atoms from their crystalline lattice sites. This displacement damage (frequently measured in numbers of displacements per atom, or dpa) produces a wide range of mechanical and physical property changes that vary with neutron flux, spectrum, and temperature. These environmental parameters vary widely from one reactor system to another and also within any given system, depending upon distance from the fission or fusion events.

Successful materials development requires irradiation testing facilities that reproduce as closely as possible the thermal and neutronic environment expected in a power-producing reactor. In some instances, this can be done fairly readily. For example, in the case of materials development for Fast Breeder Reactors (FBRs), a test reactor such as the Fast Flux Test Facility (FFTF) at Hanford provides a testing environment that closely resembles that expected for a commercial reactor. Furthermore, it has the capacity to go beyond materials testing and to accommodate full-scale reactor components for integrated fuel testing. On the other hand, materials development for Magnetic Fusion Energy (MFE) machines is complicated by the absence of any irradiation facilities which reproduce the neutron environment typical of a fusion reactor. In order to simulate the most important characteristics of the fusion neutron spectrum, it is necessary to use a wide range of irradiation facilities such as fast and mixed spectrum fission reactors, low power neutron sources and heavy ion accelerators.

In developing damage-resistant materials, much of the initial work is based upon information gained from irradiations carried out to fairly modest fluences. However, it eventually becomes necessary to subject materials to neutron exposures representative of a component lifetime exposure in a power producing reactor. Using existing fission reactors, lifetime scoping irradiations for breeder or fusion reactor applications take a minimum of three years. To produce new or modified materials, to conduct irradiations to a level approaching a lifetime fluence, and to conduct post-irradiation evaluation, requires a period of at least five years. This long-time cycle, the complexities of the irradiation environment, and the difficulties in testing and examining highly radioactive materials, all mitigate against rapid progress in materials development. Materials irradiation facilities in a very high flux neutron facility could significantly improve this situation, particularly for materials development programs requiring irradiations to damage levels of 100 dpa or more. Another factor to be considered in planning an advanced steady-state neutron facility is that many of the research reactor facilities currently being used for engineering materials development were constructed in the early 1960's, and will probably be phased out of operation during the next ten years. Even for programs which would not benefit from higher neutron fluxes, consideration should be given to providing new facilities that are equivalent to current facilities in terms of flux, spectrum and instrumentation.

In the following, the effects of neutron irradiation on the properties of engineering materials are briefly summarized. Neutron irradiation facilities currently used in support of U.S. materials development programs are described and the potential advantages of an advanced steady-state neutron facility to these programs are discussed.

RADIATION EFFECTS IN ENGINEERING MATERIALS

The various interactions between neutrons and the atoms comprising the structural materials give rise to a variety of mechanical, physical, and chemical property changes collectively referred to as "radiation effects". A broad review of this area of materials science has been presented by Olander [1]. Radiation effects arise from a combination of atomic displacements and neutron capture events. Neutrons with sufficient energy displace atoms from their atomic site. These initially-displaced atoms, or primary knockon atoms (PKAs), subsequently create damage in the lattice in the form of displacement cascades. A variety of nuclear reactions [(n,p),(n,α),(n,2n), etc.] introduce solid and gaseous transmutation products which may strongly affect materials properties. Of particular importance is the formation of helium from (n,α) reactions. Many of the reaction products are radioactive isotopes with very long half-lives, which give rise to problems with the safe disposal of reactor components at end-of-life.

The point defects which survive the collapse of the displacement cascades, diffuse through the host lattice. A major fraction of this population is annihiliated through mutual recombination of vacancies and interstitials. Those avoiding recombination either migrate to pre-existing sinks such as grain boundaries, interfaces, and dislocations, or cluster to form a variety of extended defects. For example small cavities may develop via the clustering of vacancies, and interstitials may aggregate to form extra platelets of atoms, i.e. interstitial dislocation loops. Accompanying the fluxes of point defects are flows of solute atoms towards or away from the various internal sinks. This phenomenon of radiation-induced segregation results in local regions becoming highly enriched or depleted in selected solute elements. This local un-mixing often results in major changes in alloy precipitation behavior.

The principal radiation effects which influence the dimensional stability of engineering materials are void swelling and radiation creep. At sufficiently high fluences, small cavities, initially stabilized by transmutation-produced helium, transform into continuously growing voids. This growth is driven by the bias in the microstructural system arising from the preferential attraction between interstitials and dislocations. At temperatures in the range 0.3 to 0.6 of the melting point (T_m), this phenomena can give rise to dimensional changes of the order of tens of percent. Radiation creep, or the gradual plastic deformation of a material subjected to stress, is strongly coupled to void swelling. Several mechanisms of irradiation creep have been proposed. These include the preferred absorption of point defects at dislocations or interstitial loops that are favorably oriented with respect to the stress, and dislocation glide enabled by dislocation climb resulting from point defect absorption.

Neutron irradiations can also induce major changes in mechanical properties. At low to moderate temperatures (0.30-0.45 T_m), the yield stress of materials such as ferritic and austenitic stainless steels is raised substantially by the formation of small defect clusters, loops, and precipitates. Such hardening is invariably accompanied by a reduction in tensile ductility. In the case of the ferritic steels, radiation hardening may lead to a transition to a cleavage mode of fracture and a dramatic reduction in toughness. At higher irradiation temperatures, grain boundaries may become weakened by some combination of solute segregation, precipitation, and helium bubble formation. Such phenomena may significantly reduce high-temperature strength and ductility and consequently impose limits on the allowable design stresses. In certain radiation environments, nuclear transmutation rates may be high enough to have a significant effect on critical physical properties. For example, the thermal and electrical conductivities of copper and copper alloys used for magnet components and for high-heat-flux components in fusion reactors are significantly degraded by the formation of nickel and zinc impurity atoms from transmutation reactions with certain copper isotopes. In the relatively new field of radiation effects in ceramic materials, changes in thermal conductivity, strength, dielectric properties, and RF transmissivity due to neutron irradiation are of growing interest and concern.

In recent years, spending on radiation effects work has been running at ~ $25M annually in the United States, with Japan, the European Community, and the USSR also supporting fairly large programs. The primary support for irradiation experiments on engineering materials in the United States has come from the major reactor programs - namely, Fast Breeder Reactor (FBR), Light Water Reactor (LWR), Gas Cooled Reactor (GCR), Space Power Reactor (SPR), and Magnetic Fusion Energy (MFE). The level of funding for materials irradiations in support of these programs fluctuates with congressional perception of the energy future of the United States. Currently, radiation effects work in support of the LWR and GCR programs is at a low ebb and

FBR-related work is declining steadily. On the other hand, MFE programs in the United States and Japan currently support vigorous materials irradiation studies and there is a growing interest in radiation effects within the SPR programs. It is impossible to predict with any certainty where the major support for irradiation programs will come from, say ten years from now. However, in planning for future irradiation facilities, it would be prudent to consider the requirements of each of the current programs.

A reference core design for a very high flux reactor at the proposed Center for Nuclear Research (CNR) has been proposed by F.C. Difilippo et al. [2]. In the following sections, the potential benefits of the CNR reference reactor design, to materials programs, are discussed. This design envisages materials irradiation facilities situated in the Inter-Fuel Zone (IFZ) and also in the Epithermal Peak Region (EPR) of the reactor. The former is characterized by a fast:thermal neutron flux ratio of ~ 50:1. The fast flux of ~ 3.8×10^{19} $m^{-2}s^{-1}$, coupled with the high duty factor (85%) would enable damage levels approaching 100 dpa/year to be achieved with the reactor operating at 135 MW. The EPR is characterized by a fast:thermal neutron flux ratio of ~ 0.3 and a fast flux of ~ 0.9×10^{19} $m^{-2}s^{-1}$. In this region the displacement damage rate would be ~ 25 dpa/year. The neutron fluxes in these positions are compared with those of current HFIR and FFTF facilities in Table 1. The following section describes the types of irradiation

TABLE 1

FLUX COMPARISONS FOR VARIOUS MATERIALS IRRADIATION FACILITIES

	FLUX 10^{19} $m^{-2}s^{-1}$				
	CNR*		HFIR		FFTF
	IFZ	EPZ	TARGET	RB	MOTA
FAST	3.8	0.9	1.4	0.5	3.0
EPITHERMAL	0.4	1.5	0.6	0.4	--
THERMAL	0.08	3.0	3.0	1.7	--

*Flux values for the ORNL reference core design operating at 135 MW.

facilities and techniques currently used by each major U.S. program and summarizes the potential benefits of irradiation facilities in the CNR reactor.

MAGNETIC FUSION ENERGY PROGRAM

The successful development of a major energy source based upon the fusion of light atoms will require solutions to a diverse range of materials problems particularly for reactor components subjected to intense fluxes of neutrons. These components include not only the first wall and associated blanket structure but also limiters, divertors, R.F. launchers, armor, insulators, dielectric breaks, etc. Materials currently under development for those varied applications include austenitic and ferritic stainless steels, vanadium alloys, high strength copper alloys, graphite, BeO, and Al_2O_3. An overall review of this subject may be found in reference [3].

A possible fusion reactor spectrum is shown in Fig. 1 together with spectra from fast breeder and mixed spectrum reactors. This information is taken from an earlier review of irradiation facilities for fusion materials development [4]. Some 80% of the energy released in the (D,T) fusion reaction is carried into the first wall and blanket structure by 14 MeV neutrons. This 14 MeV component of the fusion spectrum produces a higher energy component of the PKA recoil spectra and a higher rate of transmutations (through threshold (n,α) and (n,p) reactions), compared to similar neutron fluxes in a fission reactor. From the point of view of materials property changes, the most significant impact of the 14 MeV neutrons is thought to be the enhanced production of helium from (n,α) reactions. The unique recoil energy distribution and production of solid transmutants are believed to be of secondary importance. The current irradiation strategy therefore is to utilize the fission reactors to simulate the displacement damage and helium generation rates expected in a fusion environment. Details of the irradiation facilities in current use are presented in Table 2.

Typically, the ratio of helium production rate to displacement damage rate (He:dpa ratio) is in the range 5-15 for structural materials in a fusion environment. The principal method of varying this ratio during fission reactor irradiations is to utilize the two-step Ni reaction, namely;

$$^{58}Ni + n \rightarrow {}^{59}Ni + \gamma \; ; \quad {}^{59}Ni + n \rightarrow {}^{56}Fe + \alpha$$

These reactions have large thermal neutron cross-sections and in addition there is a large resonance in the ^{59}Ni cross-section at ~ 203 eV. Although other reactions could be used (e.g. the $^{10}B(n,\alpha)^{6}L_{1}$ reaction), the use of Ni is metallurgically more acceptable. For alloys, which normally contain nickel, (e.g. the austenitic stainless steels), the technique of isotopic tailoring may be used to control the rate of helium production during irradiation in a mixed-spectrum reactor. The naturally occurring isotope ^{60}Ni has a very low ($n\alpha$) cross-section and therefore the helium production rate can be adjusted over a wide range by altering the proportions of ^{60}Ni and ^{58}Ni during alloy manufacture. An example of the range of helium production rates which can be achieved by this technique in a steel containing 16 wt% Ni is shown in Fig. 2. The initial transient in helium production rate can be eliminated if necessary by the addition of small quantities of the radioactive isotope ^{59}Ni during alloy manufacture. The helium production rate can also be manipulated in alloys which normally do

TABLE 2

CHARACTERISTICS OF IRRADIATION FACILITIES IN CURRENT USE BY VARIOUS U.S. PROGRAMS

EXPERIMENTAL PARAMETERS	LIGHT WATER REACTOR PROGRAM	GAS COOLED REACTOR PROGRAM	FAST BREEDER & SPACE REACTOR PROGRAMS	MAGNETIC FUSION ENERGY PROGRAM	
U.S. Reactors	Various Research Reactors ORR(Poolside)	ORR, HFIR	FFTF	ORR, HFIR	FFTF
Spectrum	Mixed	Mixed	Fast	Mixed	Fast
Flux (n/m^2) ($E > 0.1$ MeV)	$(5\text{-}50) \times 10^{16}$	$(5\text{-}10) \times 10^{18}$	$(1\text{-}3) \times 10^{19}$	$(0.3\text{-}1.5) \times 10^{19}$	$(1\text{-}3) \times 10^{19}$
Thermal Flux (n/m^2)	$(5\text{-}50 \times 10^{16}$	$(5\text{-}10) \times 10^{18}$	--	$(0.2\text{-}3) \times 10^{19}$	--
Irradiation Time (days)	100 - 500	30 - 300	200 - 800	50 - 1000	200 - 800
Capsule Diameter (cm)	2 - 25	2 - 7	2.5	1.5 - 5.0	2.5
Instrumentation	Temp. Monitoring Temp. Control	Tem. Monitoring* Temp. Control* Fission Gas Detection*	Temp. Measurement Temp. Control	Temp. Measurement Temp. Control*	Temp. Measurement Temp. Control
Temperature Range (°C)	100 - 300	200 - 1250	380 - 1500	60 - 750	380 - 750
Materials	Low Alloy Ferritic Steels	Coated Fuel Particles Graphites	Austenitic & Ferritic Stainless Steels Refractory Metals & Alloys	Austenitic & Ferritic Stainless Steels Vanadium Alloys Copper Alloys Ceramics Tritium Breeding Materials	

*Only in ORR

not contain nickel by doping with small quantities of ^{58}Ni, provided the metallurgical stability of the alloy is not seriously perturbed. Isotopic tailoring with ^{59}Ni may be used to control helium generation in fast reactor irradiations. However, in order to utilize the 203 eV resonance, irradiations must be carried out in regions of the core where the displacement damage rate is low.

Another important technique, applicable to nickel-bearing alloys is that of spectral tailoring. In this technique, the local spectrum in a mixed-spectrum reactor is modified by surrounding the experiment with absorber shields. An example of helium production in a spectral tailoring experiment in the ORR is shown in Fig. 3. A less satisfactory approach is to introduce helium into materials prior to irradiation either by cyclotron injection or by using the "tritium trick". The latter technique is currently being utilized to explore helium effects in vanadium alloys [5]. Tritium is diffused into the material and allowed to decay to the required fraction of ^{3}He before pumping out the residual tritium. The doped material is then irradiated in a hard neutron spectrum.

An important aspect of materials irradiation experiments is the miniaturization of physical and mechanical property measurement techniques, in order to make the most effective use of the available irradiation space. Specimens with dimensions of the order of a few millimeters are being successfully utilized for swelling, creep, ductility, strength and crack resistance measurements [6], [7].

Although these irradiation techniques do not provide a complete simulation of the fusion environment, they do allow progress to be made both in understanding high temperature radiation damage phenomena, and in improving the radiation damage resistance of certain classes of materials.

Exploration of the synergistic effects of the solid and gaseous transmutation products and the primary displacement damage produced by 14 MeV neutrons requires the construction of a powerful 14 MeV neutron source, such as the Fusion-Materials Irradiation Test Facility (FMIT) [8]. However, a disadvantage of machines such as FMIT is the limited experimental volume within which the flux is sufficiently high, and flux and temperature gradients are acceptable. Until such time as a fusion-based materials test facility is constructed, the materials development strategy for fusion will require both a 14 MeV neutron source and high quality fission reactor facilities. The proposed ORNL reference core design for CNR could fulfill this latter function and provide several important advantages over existing fission reactor facilities as discussed below:

a) <u>High fast neutron flux</u>

The combination of a high flux hard spectrum in the IFZ coupled with a high duty factor (~ 85%) would allow damage levels of ~ 100 dpa to be reached in one year instead of the three years currently required in FFTF or HFIR. Such a facility could halve the time cycle required to prepare alloy modifications and assess their performance at high damage levels. It would also allow, for the first time, an exploration of materials response to damage levels beyond 100 dpa.

b) <u>Hard and mixed spectra</u>

The mixed spectrum facilities in the EPR would be ideally suited for larger diameter spectrally tailored experiments for the irradiation of isotopically tailored alloys. Damage accumulations would be at the rate of ~ 25 dpa/year, i.e. double the rate currently achieved in a HFIR removable beryllium (RB) position. For materials containing appropriate levels of ^{58}Ni, a more rapid damage rate could be achieved by alternately irradiating the same capsule in the EPR and IFZ positions. A period of rapid helium buildup followed by a period of rapid damage accumulation could be used to produce, on average, the correct He:dpa ratio.

c) Coolant temperature

The use of water as the reactor coolant allows irradiation experiments to be carried out over the complete range of interest to fusion materials applications, i.e. from 100°C up to 750°C; the minimum temperature attainable in a liquid metal cooled fast reactor is ~ 380°C.

d) Flexibility

The relatively easy access to experimental positions and a fuel cycle of the order of ~ 20 days allows experimenters maximum flexibility in scheduling experiments. This is particularly important for basic experiments where incremental doses in the range 1-10 dpa are frequently needed.

FAST BREEDER REACTOR PROGRAM

Materials for core components such as fuel cladding and flow ducts are subjected to high levels of displacement damage. For example, for a three-year component lifetime, structural materials will have to withstand at least 100 dpa. The primary objective of materials testing for breeder reactor components is to expose a wide variety of materials, in a wide range of specimen geometries, to fast-neutron fluences of this order, at temperatures ranging from 380° to 750°C. The Fast Flux Test Facility (FFTF) at Hanford was built for materials and component testing. The Materials Open Test Assembly (MOTA) was specifically designed for irradiating large numbers of diverse specimens in a continuously monitored, controlled temperature environment to high levels of neutron fluence at damage rates similar to those expected in a large power producing reactor. Austenitic and ferritic stainless steels are the materials of principal interest. Post-irradiation measurements of a wide range of properties are required (e.g., swelling, irradiation creep, tensile, fracture toughness, Charpy impact, and microstructural evolution). In-reactor measurements of the rupture life of pressurized tubes are also carried out. Irradiation experiments in MOTA accumulate displacement damage at a rate of ~ 30 dpa/year. The capability of accumulating ~ 100 dpa/year in the IFZ of the CNR reactor would allow end-of-life fluences for breeder reactor core materials to be achieved at an accelerated rate. Using small scale testing techniques, the swelling behavior and mechanical properties of large numbers of alloy variants exposed to very high damage levels could be assessed in a relatively short time. Subsequently the properties of the more promising materials would be explored in MOTA using a wider range of test specimens. Used in this complementary fashion, CNR irradiation facilities could be a major asset to FBR materials programs.

SPACE POWER REACTOR PROGRAMS

Present United States concepts for space power reactors (SPRs) center around fast reactors operating at very high temperatures (> 1100°C) and utilizing structural alloys based upon refractory metals (Nb, Mo, W, Rh).

End-of-life fluence levels are modest (10-20 dpa) and the MOTA provides an ideal facility for irradiation testing of these materials under neutronic conditions very similar to those projected for a space power reactor. Using liquid-metal-filled capsules surrounded by gas gaps with variable-gas composition, specimen temperatures of 1000 to 1350°C can be achieved. Future concepts may raise the power of space reactors into the multi-megawatt range, in which case core components may be expected to absorb damage levels of the order of ~ 100 dpa and maintain their dimensional and structural integrity for very long lifetimes (> 5 years). As long as facilities are available in the FFTF, high flux facilities in the CNR reactor would provide no additional benefits to SPR programs involving low power concepts. However the ability to achieve end-of-life damage levels on an accelerated basis could be of considerable benefit to multi-megawatt SPR materials development programs. As in the case of materials development for FBRs, rapid alloy screening based on a limited range of properties could be achieved before proceeding to more extensive testing in MOTA.

GAS-COOLED REACTOR PROGRAMS

The primary goal of irradiation experiments in this area is to assess the performance of new experimental fuels, the integrity of fuel particle coatings, and to determine the effects of irradiation on the mechanical and physical properties of graphite. The characteristics of currently used facilities are summarized in Table 2. Facilities in mixed spectrum reactors are used to create fissions in the fuel particles and neutron damage in the particle coatings. A high level of instrumentation is required to monitor and control temperature and also to monitor fission product release into a sweep gas.

The fluxes currently available in the RB positions in HFIR and in the ORR already allow irradiation testing at a sufficiently accelerated rate. There are no advantages to be gained by providing facilities with higher fluxes, such as in the IFZ in the proposed CNR reactor. However, irradiation facilities in the EPR would provide an excellent replacement for currently used facilities in HFIR and ORR should these reactors eventually be phased out.

LIGHT-WATER REACTOR PROGRAMS

The primary radiation effects problem in this area involves the effects of long-term neutron exposure on the mechanical properties of the ferritic steels used to construct pressure vessels for light-water reactors. The characteristics of currently used irradiation facilities are summarized in Table 2. To reproduce the damage conditions experienced by a light-water reactor pressure vessel, mixed spectrum reactors are used with sufficient fast neutron flux to simulate end-of-life exposure conditions at an accelerated rate if necessary. Large capacity facilities are required to accommodate fairly massive test specimens and temperature measurement and control instrumentation is essential with most irradiations being conducted in a narrow temperature range around 280°C. Post-irradiation property measurements include fracture toughness, Charpy impact, hardness and tensile properties.

The fluxes available in current low power research reactors already allow irradiation testing at a sufficiently accelerated rate of damage accumulation. For LWR materials programs, there are no advantages to be gained by providing facilities with higher fluxes. The requirements of this program will probably be adequately met in the future by various low-power research reactors.

SUMMARY

The development of materials with adequate resistance to radiation-induced property changes requires irradiation facilities which simulate the principal spectral characteristics of power-producing reactor systems. Considerable savings in time and money can be effected by carrying out irradiations at an accelerated rate, provided the dose rate is not high enough to affect microstructural development. As far as U.S. gas-cooled reactor and light water reactor materials programs are concerned, existing research reactors already have the capability for carrying out irradiations at a sufficiently accelerated rate. For these programs, there are no advantages to be gained from irradiation facilities in a CNR reactor. On the other hand, materials programs for fast breeder and space power reactors would benefit substantially from the capability to achieve high levels of radiation damage in a hard spectrum at 2-3 times the rate currently achievable in breeder reactors. The proposed ORNL reference core design for a CNR reactor would be an outstanding asset to fusion materials programs. A set of well instrumented, accessible positions in both hard and mixed spectral regions would provide un-paralleled facilities for isotopic and spectral tailoring experiments and for exploring very high damage levels at an accelerated rate.

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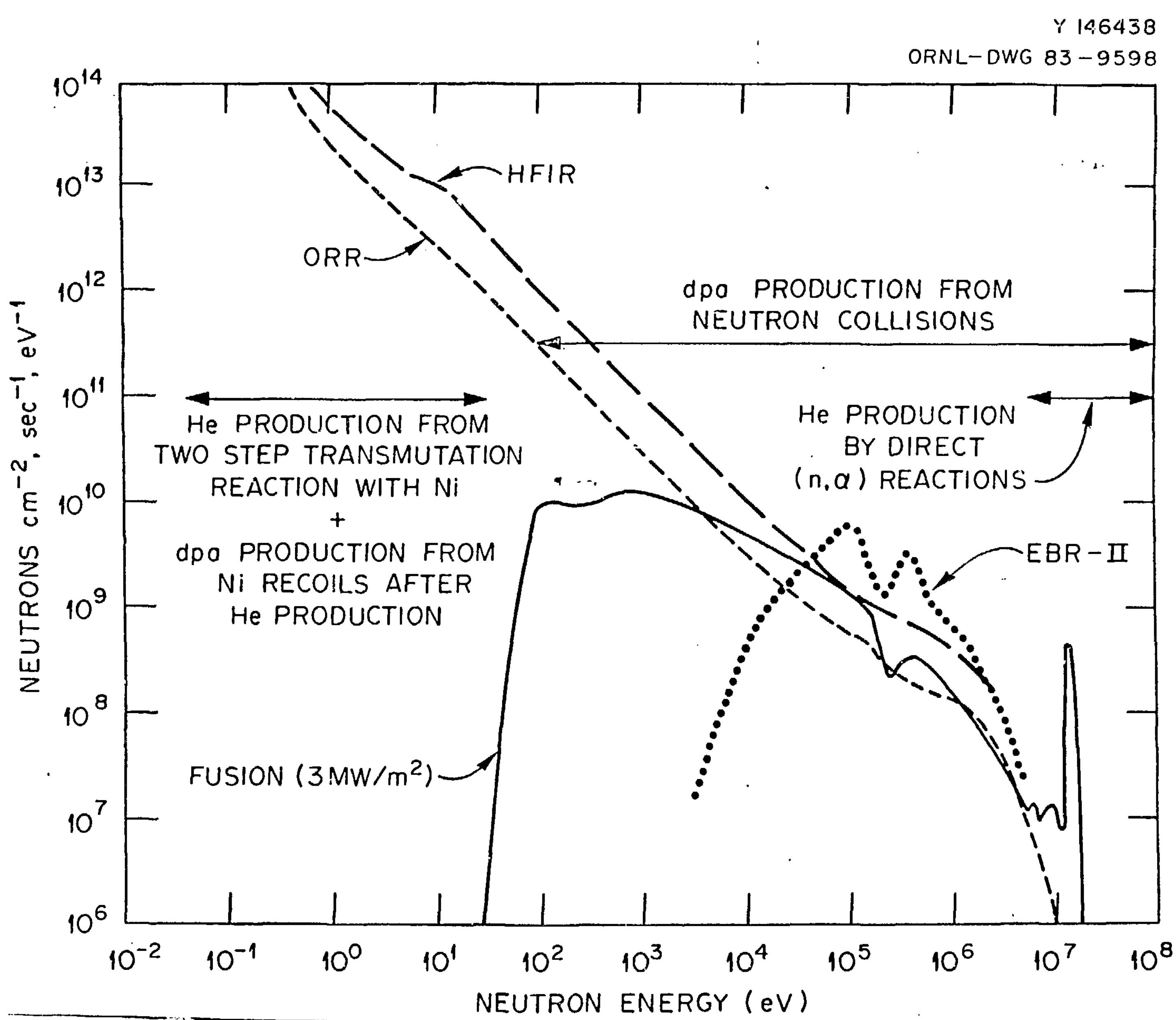

Fig. 1. A comparison of neutron spectra for a fusion reactor and various fission reactors.

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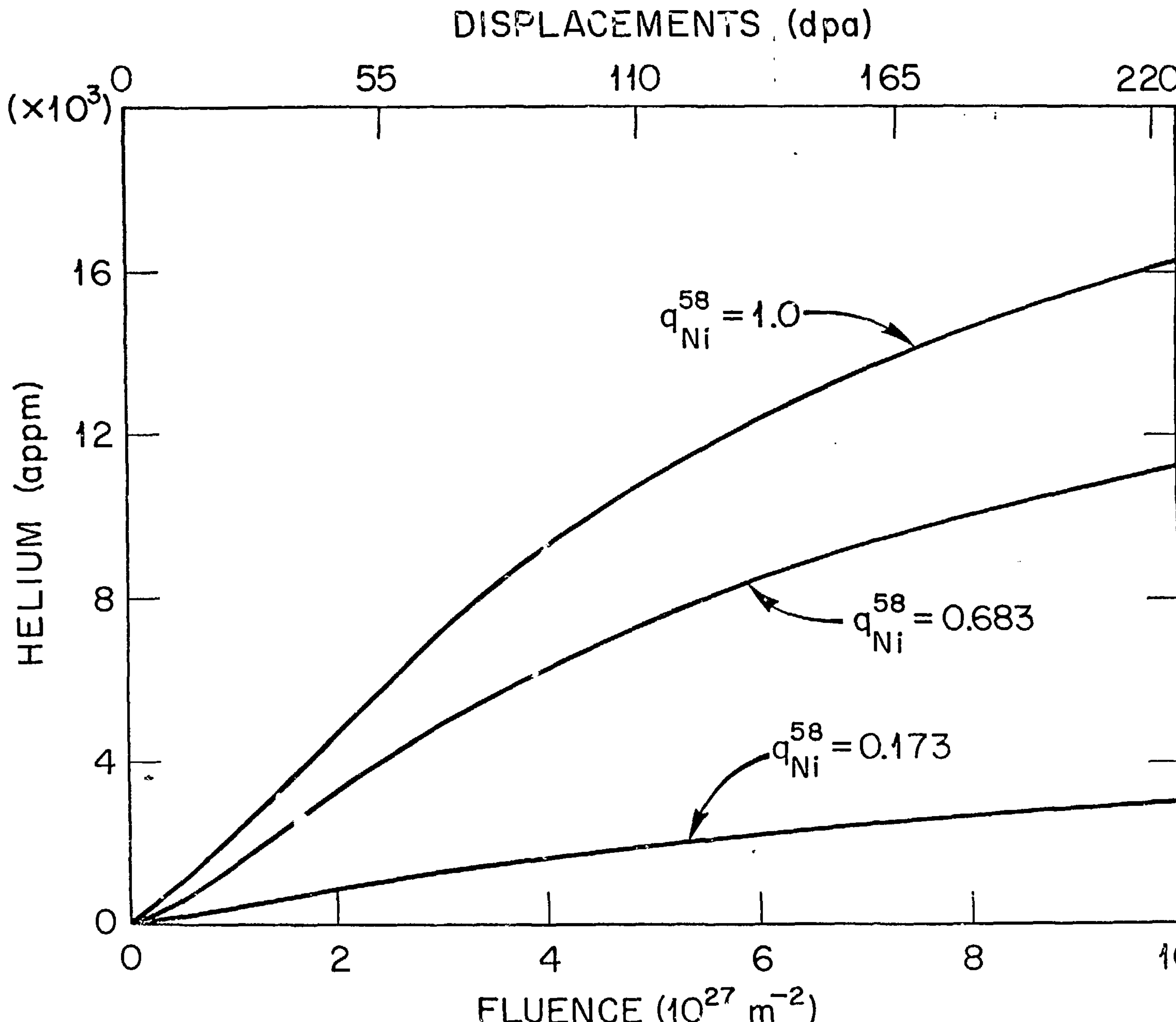

Fig. 2. Helium production as a function of displacement damage in an austenitic stainless steel containing 16 wt % Ni during irradiation in the HFIR target region. The helium generation rate is varied by altering the initial factions of ^{58}Ni, (q_{Ni}^{58}).

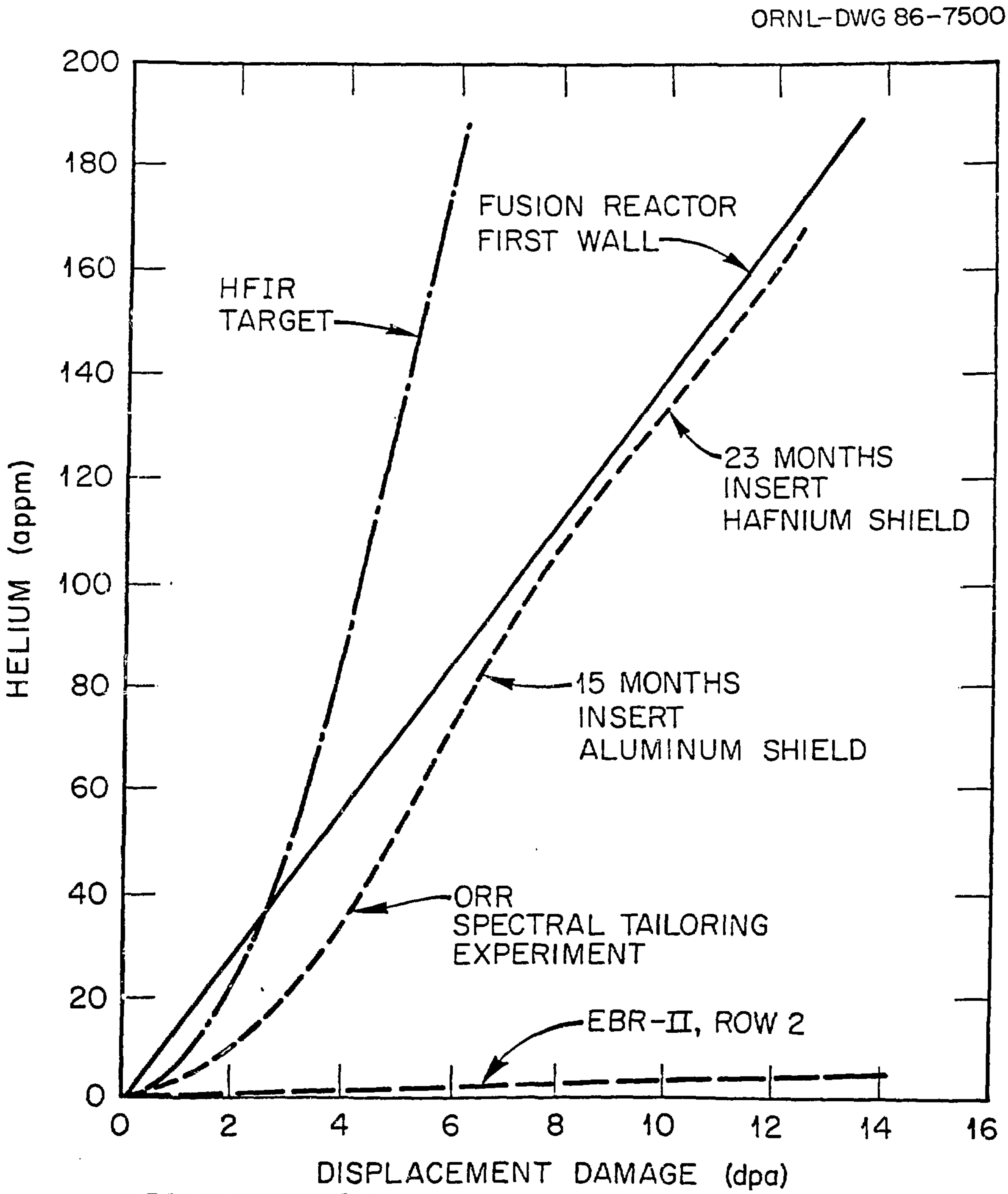

Fig. 3. Helium production as a function of displacement damage for AISI 316 stainless steel. The He:dpa ratio for a fusion reactor first wall is intermediate between those occurring during irradiations in HFIR and in EBR-II. A spectral tailoring experiment in the ORR produces a good simulation of the fusion conditions.